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RADON REMOVAL TECHNIQUES FOR SMALL COMMUNITY PUBLIC WATER SUPPLIES

by

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16. ABSTRACT

This report presents the results of an evaluation, performed by the University of New Hampshire--Environmental Research Group (ERG), of radon removal in small community water supplies using full-scale granular activated carbon adsorption, diffused bubble aeration and packed tower aeration. Various low technology alternatives, such as loss in a distribution system and addition of coarse bubble aeration to a pilot-scale atmospheric storage tank were also evaluated. The report discusses each of the treatment alternatives with respect to their radon removal efficiency, potential problems (i.e., waste disposal, radiation exposure and intermedia pollution), and economics in smal community applications. In addition, several sampling methods, storage times, scintillation cocktails and extraction procedures currently used in the liquid scintillation technique for analysis of radon in water were compared.

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FOREWORD

Today's rapidly developing and changing technologies and industrial products and practices frequently carry with them the increased generation of materials that, if improperly dealt with, can threaten both public health and the environment. The U.S. Environmental Protection Agency is charged by Congress with protecting the Nation's land, air, and water resources. Under a mandate of national environmental laws, the agency strives to formulate and implement actions leading to a compatible balance between human activities and the ability of natural systems to support and nurture life. These laws direct the EPA to perform research to define our environmental problems, measure the impacts, and search for solutions.

The Risk Reduction Engineering Laboratory is responsible for planning, implementing, and managing research, development, and demonstration programs to provide an authoritative, defensible engineering basis in support of the policies, programs, and regulations of the EPA with respect to drinking water, wastewater, pesticides, toxic substances, solid and hazardous wastes, and superfund-related activities. This publication is one of the products of that research and provides a vital communication link between the researcher and the user community.

This report presents the results of an evaluation, performed by the University of New Hampshire - Environmental Research Group (ERG), of radon removal in small community water supplies using full-scale granular activated carbon adsorption, diffused bubble aeration and packed tower aeration. Various low technology alternatives, such as loss in a distribution system and addition of coarse bubble aeration to a pilot-scale atmospheric storage tank were also evaluated. The report discusses each of the treatment alternatives with respect to their radon removal efficiency, potential problems (i.e., waste disposal, radiation exposure and intermedia pollution), and economics in small community applications. In addition, several sampling methods, storage times, scintillation cocktails and extraction procedures currently used in the liquid scintillation technique for analysis of radon in water were compared.

E. Timothy Oppelt, Director
Risk Reduction Engineering Laboratory

ABSTRACT

Radon is one of the naturally-occurring radionuclides which will be regulated as part of the Safe Drinking Water Act Amendments of 1986. The proposed rule must contain recommendations with respect to Best Available Technologies (BAT) and analytical methods. The purpose of this project was to evaluate the performance of full-scale granular activated carbon (GAC) and diffused bubble and packed tower aeration systems treating small community groundwater supplies containing radon. In addition, several low technology alternatives were evaluated including radon loss in a distribution system and pilot-scale tests of modifications to atmospheric storage tanks, such as coarse bubble aeration. Total production costs are provided for each treatment system evaluated, however, these are highly dependent on process equipment and percent removal required, which are site specific. The liquid scintillation analytical technique for radon was also evaluated with respect to sampling methods, storage times, extraction methods and cocktail selection.

GAC systems were operated at two small communities in New Hampshire. During continuous operation at the Mont Vernon site, the effluent radon activity averaged 24,653 ± 17,016 pCi/L (influent activity = 210,491 ± 41,384 pCi/L; flowrate = 36 ± 12 m^3/day). The GAC effluent activity at the Amherst site averaged 12,000 ± 5,900 pCi/L (influent activity = 41,800 ± 15,200 pCi/L; flowrate = 61 ± 5 m^3/day). The data indicated that the GAC units retained uranium, radium and lead. Concentration of these radionuclides appeared to be related to raw water quality and their retention could lead to classification of the GAC as a low level radioactive waste in some states. The GAC units also accumulated iron, manganese, particulates and microorganisms which may necessitate periodic backwashing or pretreatment to prevent significant headloss development. Gamma/beta emissions measured at the surface of the GAC units were substantially greater than background and shielding may be required to lower emissions to acceptable levels.

For the diffused bubble system operating at air:water (A:W) ratios $\geq$ 5:1 at high water flowrates (0.10 ± 0.0019 m^3/min; influent radon = 65,487 ± 5,657 pCi/L) and low water flowrates (0.047 ± 0.00053 m^3/day, influent radon = 78,385 ± 6,120 pCi/L), the overall radon removal efficiency ranged from 90.0 to >99.6%. The removal efficiencies for the packed tower ranged from 92.7 to 99.8% over the range of air flowrates tested in spite of variation in water flowrate, influent radon activity and packing type. Off gas radon activities were 10^4 to 10^5 times higher than ambient air levels for both aeration systems and the impact of the discharge on air quality should be considered. Precipitation of iron and manganese could result in operational problems in both systems and may necessitate pretreatment.

Radon loss during water transport in a distribution system (497 m, hydraulic retention = $\leq$ 17.9 min) was not effective in radon removal. In pilot-scale testing, atmospheric storage modified by free fall and spray nozzle entry and coarse bubble aeration achieved radon removals of 50 to 96%, but the system may have problems with off-gas emissions and/or iron and manganese precipitation.

When designing any system to remove radon from a small community water supply, it is imperative to have good data on water flowrates and influent radon activities. As observed in this study, variations in these parameters may be substantial and will, if underestimated, lead to inadequate system design and effluent radon activities which exceed the design goal. Since GAC adsorption-decay constants and aeration mass transfer coefficients are system specific, extrapolations of performance data obtained at one site should not be made to systems with (i) other design configurations, (ii) low influent radon activities and/or (iii) a requirement to meet a more stringent MCL. Pilot-scale studies should be conducted at each site to obtain the required design information.

Water samples collected using the direct syringe technique had higher radon activities than those collected in VOA bottles, but widespread use of this technique may be limited by costs, problems associated with distribution of syringes and the difficulty in collecting samples devoid of air bubbles. Sample storage in VOA bottles was affected by radioactive decay and leakage (losses up to 20% in 21 days). The OptiFluor O scintillation cocktail yielded significantly higher count rates than a toluene-based cocktail and was less expensive than the toluene and mineral oil-based cocktails. The extraction experiment data suggested that radon in a radium-226 standard is continuously transferred to the cocktail phase regardless of shaking and therefore, the extraction procedure should not be used to calculate the efficiency factor.

This report was submitted in partial fulfillment of Cooperative Research Agreement CR812602-01-0 by the Environmental Research Group of the University of New Hampshire under the sponsorship of the U.S. Environmental Protection Agency in conjunction with the State of New Hampshire Department of Environmental Services. This report covers the period January 1986 to December 1988 and work was completed as of June 1989.

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LIST OF ABBREVIATIONS AND SYMBOLS

ABBREVIATIONS

ANOVA -- analysis of variance
A:W -- air to water ratio
CCI -- construction cost index
CFU -- colony forming units
DDI -- double deionized water
DI -- deionized water
DO -- dissolved oxygen
ENR -- Engineering News Record
EBCT -- empty bed contact time
FOB -- free on board
GAC -- granular activated carbon
GM -- Geiger Müeller
HTU -- height of transfer unit
LSC -- liquid scintillation counter
MCL -- maximum contaminant level
NTU -- nephelometric turbidity unit or number of transfer units
O&M -- operation and maintenance
PSNH -- Public Service Company of New Hampshire
SPC -- standard plate count
StT -- Student's t test
TDC -- total direct costs
VOA -- volatile organics analysis

SYMBOLS

α -- level of statistical significance
β -- contaminant diffusivity
C_A -- activity of radon in the air
C_{eff} -- effluent radon activity in water
C_f -- packing factor
C_G -- activity of radon in the gas phase
C_{inf} -- raw water (influent) radon activity
C_i -- activity of radon in the liquid phase
C_m -- density of the liquid
d_b -- bubble diameter
D_o -- oxygen diffusivity
D_w -- molecular diffusion coefficient

E -- efficiency factor
G -- air loading rate
H -- Henry's constant
H_L -- sidewall depth
K_La -- global mass transfer coefficient
K_{ss} -- steady state adsorption-decay constant
L -- liquid loading rate
mR -- milliRoentgen
mREM -- milliRoentgen Equivalent Man
P_t -- ambient pressure
ρ_w -- density of water
Q_g -- air flowrate
R -- stripping factor
$t_{1/2}$ -- radionuclide's half-life
μ_m -- viscosity of water
V_c -- liquid volume of diffused bubble tank
z -- packing height

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SECTION 1

INTRODUCTION

As part of the 1986 amendments to the Safe Drinking Water Act, the United States Environmental Protection Agency (EPA) will propose a rule for Maximum Contaminant Level Goals (MCLGs) and National Primary Drinking Water Regulations (NPDWR) including Maximum Contaminant Levels (MCLs) for radionuclides in drinking water (Federal Register, 1986). One of the radionuclides which will be regulated under the proposed rule is radon-222* (hereafter referenced as radon in the text). In a recent status report, EPA (1989) indicated it is considering setting the MCL for radon in the range 200 to 2,000 pCi/L. There have been several studies of the distribution of radon in groundwater supplies in the U.S. (Hess et al., 1985; Longtin, 1988; Dixon and Lee, 1988; Vitz, 1988) all of which indicate that there will be a large number of point-of-entry and public water supplies impacted by an MCL in that range. Many of the public water supplies affected will be those serving small communities (<76 m^3/day).

The rule will also contain recommendations with respect to Best Available Technologies (BATs) and analytical methods. Three conventional water treatment technologies - granular activated carbon adsorption (GAC), diffused bubble aeration, and packed tower aeration - have been used to remove radon from drinking water. Most of the studies in the U.S. on GAC have been performed by Lowry and associates (Lowry and Brandow, 1981, 1985; Lowry, 1985; Lowry et al., 1987, Lowry and Lowry, 1987; Lowry and Lowry, 1988; Lowry et al; 1988), though Dixon and Lee (1987) reported some radon removal in GAC units designed for organics treatment. The GAC process, which has been used to treat point-of-entry and small community water supplies, relies on the ability of radon to adsorb to the carbon. One unique aspect of the radon removal process is the fact that the bed is naturally regenerated as radon decays (Lowry and Brandow, 1985). As a result, the breakthrough/exhaustion profile typically seen when GAC is treating other conservative contaminants is not exhibited during radon removal.

The aeration methods are predicated on the fact that radon is a highly volatile gas with a relatively high Henry's constant (2.80 atm $\cdot$ $m^3 H_2O/m^3$ air at 10°C) and thus can be easily transferred from water to air. Aeration methods have been used in both point-of-entry and community public water supplies (Perkins and Hewett, 1988; Cummins, 1987; Dixon and Lee, 1987; Lowry et. al., 1987; Hoather and Rackham, 1962; Castren, 1977; Lowry and Brandow, 1981).

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* Though there are many radioactive isotopes of radon, radon-222 is the most common.

In addition, Dixon and Lee (1987) have examined the potential of alternative technologies (for example, cascading tray aerators used for iron and manganese removal and loss in standpipes) which use simple devices often already in place and adapt them to radon treatment.

The analytical methods currently being examined by EPA (1989) for the proposed radon rule include liquid scintillation counting (Pritchard and Gesell, 1977; EPA, 1978), and the Lucas cell method (Lucas, 1957). These techniques have been used extensively and have been found to be equivalent in interlaboratory studies conducted for EPA (Whittaker et al., 1987).

RESEARCH OBJECTIVES

The purpose of this EPA Cooperative Agreement was to evaluate the performance of full-scale GAC and diffused bubble and packed tower aeration systems treating small community groundwater supplies containing radon. The water supplies contained 40,000 to 250,000 pCi/L of radon at flowrates ranging from 25 to 61 m^3/day. In addition, several low technology alternatives were evaluated including radon loss in a distribution system and pilot-scale tests of modifications to atmospheric storage tanks such as coarse bubble aeration. Each of these systems (GAC, diffused bubble aeration, packed tower aeration and the low technology alternatives) were evaluated with respect to three primary factors: radon removal efficiency, potential problems (i.e., waste disposal, radiation exposure, intermedia pollution) and economics. It was not the purpose of the study to develop design equations for these systems, but rather to evaluate their operation at small community facilities.

Another aspect of the study was the evaluation of the liquid scintillation counting technique used in radon analysis. This technique has been used with several scintillation cocktails, extraction techniques, sampling methods, and storage times. These aspects of the technique were studied to determine whether the various modifications available impact the results obtained with the method.

The specific objectives of the study were to:

- Evaluate full-scale GAC systems operating at two small communities in New Hampshire, monitoring them for changes in radon removal, radiation emissions, general water quality parameters (e.g., pH, iron, turbidity, microbial numbers);

- Conduct several special monitoring events of the GAC systems to assess the impact of diurnal variations in water flowrate and raw water quality, high water flowrate and backwashing on GAC performance;

- Core the GAC after several months of operation to determine if iron, manganese, microorganisms and/or radionuclides were accumulating in the units;
- Evaluate full-scale diffused bubble and packed tower aeration systems operating in small communities in New Hampshire, monitoring them for radon removal, general water quality parameters and off-gas emissions of radon;
- Operate the aeration systems over a range of volumetric air to water (A:W) ratios at two water flowrates to determine the effect of these parameters on radon removal;
- Evaluate three randomly packed plastic media with respect to radon removal efficiency in the tower aeration system;
- Evaluate the efficiency of radon removal in a small community distribution system;
- Evaluate the radon removal efficiency of several low technology modifications (e.g., free fall vs. bottom entry, spray nozzle entry, venturi entry, coarse bubble aeration retrofit) to an existing small community atmospheric storage tank;
- Assess the effect of sampling techniques (free fall, hose connector, direct syringe collection, VOA bottle collection), storage time, scintillation cocktail, and extraction via shaking on the liquid scintillation analytical technique for analysis of radon in water.

The report is divided into 8 sections including introduction (Section 1), conclusions and recommendations (Section 2), sampling and analytical methods (Section 3), separate sections discussing the results obtained in the evaluations of GAC (Section 4), diffused bubble aeration (Section 5), packed tower aeration (Section 6), low technology alternatives (Section 7), and the liquid scintillation analytical technique (Section 8). The treatment technology evaluation sections each contain: (i) an introduction; (ii) description of the units, experimental design and special sampling/analytical procedures; (iii) detailed presentation of the results and a discussion of their significance; and (iv) an economic analysis of the system operated.

FUNDAMENTALS OF RADIONUCLIDES

Radionuclides are different from the majority of drinking water contaminants regulated by EPA because of the instability of their nucleus and their ability to decay. This often leads to confusion resulting from the dissimilarity between the units used to describe radioactivity and those used for other water contaminants; as well as the physical meaning of these units. Therefore, a brief synopsis of some of the fundamental principles of radionuclides important to this project are discussed below.

Radon-222 is part of the uranium (4n+2) decay series (Table 1) which originates with uranium-238. Radon and its four short lived progeny (polonium-218; lead-214, bismuth-214 and polonuim-214) have some of the shortest half-lives (less than 27 min) of any of the radionuclides in the series. Radon is the only gas in the decay series and is a member of the Periodic Chart Group 0 (noble) gases including helium, argon and other chemically inert monatomic gases. Radionuclides are described in units of activity - curies - which indicate the rate at which a radioactive atom is undergoing decay (i.e., nuclear disintegrations). By convention a picocurie (pCi) means that 2.22 atoms of that radionuclide are decaying per min (disintegrations per min = dpm). A radionuclide's half-life ($t_{1/2}$) is the time interval during which 1/2 of the atoms decay ($t_{1/2} = \ln 2/\lambda$ where λ = decay constant, 0.0075 hr^{-1} for radon). For radon and its progeny the half-lives are very short varying from 3.82 days (radon-222) to 0.00016 sec (polonium-214). The actual life of an individual atom, however, can range from 0 to infinity (i.e., decay occurs randomly).

Though the units specified for radionuclides in water are in pCi/L (i.e., activity per unit volume) and their range in groundwater is 10^{0}-10^{6} pCi/L, the mass and number of atoms present is usually extremely small and can be determined using the equation: $A = N\lambda$ where A = activity (pCi or dpm) and N = number of atoms. For example, 1,000 pCi of radon (2,220 dpm) in a liter of water is only 1.76×10^{7} atoms of radon or a mass of 5.49×10^{-15}g (radon = 222g/mole) or 6,490 pg/L of water.

Therefore, when designing treatment systems to remove radon from water, though the activities may be substantial (and have important public health significance), the numbers of atoms and mass to be removed is very small. However, radon is a highly volatile gas and its concentration in water (ex. 1,000 pCi/L = 6.49×10^{-15} g/L) is still much greater than its concentration in air (0.1 pCi/L = 6.49×10^{-19}g/L).

Detection capabilities for most drinking water contaminants are in the 10^{-8} to 10^{-3} g/L range. The instrumentation used in radon analysis detects light flashes created when the radioactive particles strike fluorescing compounds and therefore, is able to measure lower concentrations (32.5×10^{-16} g/L = <500 pCi/L). The instrument's detector records the number of counts (scintillations) per minute or cpm. The efficiency of the instrument is usually not 100%, so an efficiency factor must be determined using standards to relate the cpm obtained to the dpm or pCi actually present.

Atoms of the various radionuclides in a decay series are constantly being produced and are, in turn, decaying to form other unstable nuclides. For example, radon is created by the decay of radium-226 ($t_{1/2}$ = 1602 yr) and decays to polonium-218 (radon $t_{1/2}$ = 3.82 days). Because of this continuing pattern of formation and decay, it is important to understand the concept of

TABLE 1. ABBREVIATED[*] URANIUM (4n + 2) DECAY SERIES

Chemical Notation	Historical Name	Half-life	Radiation Emitted Upon Decay[+]
$^{238}_{92}\text{U}$ ↓	Uranium	4.5×10^{9} yrs.	α
$^{234}_{90}\text{Th}$ ↓	Thorium	24.1 days	β, γ
$^{234}_{91}\text{Pa}$ ↓	Protactinium	1.17 min	β, γ
$^{234}_{92}\text{U}$ ↓	Uranium	2.47×10^{5} yrs.	α, γ
$^{230}_{90}\text{Th}$ ↓	Thorium	8.0×10^{4} yrs.	α, γ
$^{226}_{88}\text{Ra}$ ↓	Radium	1602 yrs.	α, γ
$^{222}_{86}\text{Rn}$ ↓	Radon	3.82 days	α
$^{218}_{84}\text{Po}$ ↓	Polonium	3.05 min	α
$^{214}_{82}\text{Pb}$ ↓	Lead	26.8 min	β, γ
$^{214}_{83}\text{Bi}$ ↓	Bismuth	19.7 min	B, γ
$^{214}_{84}\text{Po}$ ↓	Polonium	164 μsec	α
$^{210}_{82}\text{Pb}$ ↓	Lead	21 yrs.	β, γ
$^{210}_{83}\text{Bi}$ ↓	Bismuth	5.01 days	β
$^{210}_{84}\text{Po}$ ↓	Polonium	138.4 days	α
$^{206}_{82}\text{Pb}$	Lead	–	Stable –

[*] Protactinium-234 (Uranium Z), Astatine-218, Thallium-210 and 206 are not shown. These represent radionuclides created in <0.2% of the decays of Protactinium-234 (Uranium Xg), Polonium-218, Bismuth-214 and Bismith-210, respectively.

[+] Only listed if % emission is >0.1%.

secular equilibrium. When a radionuclide is in secular equilibrium with another radionuclide or its progeny, there is a constant amount of that radionuclide present which is a function of the rate at which it is being created and the rate of its decay. The mathematical relationships are best explained by Evans (1969). Hence, at secular equilibrium between radon and its progeny or between radium and radon, the activities of the parent and its progeny are equal. For example, when 1,000 pCi/L of radium is in secular equilibrium with radon, the activity of radon will also be 1,000 pCi/L.

SECTION 2

CONCLUSIONS AND RECOMMENDATIONS

GRANULAR ACTIVATED CARBON

1. When designing a GAC system to remove radon from a small community water supply, it is imperative to have good data on water flowrates and influent radon activities at the site. These data are major inputs into the design model for determining the volume of GAC required. As observed in this study, for small community supplies, variations in flowrate and influent activity may be substantial and will, if underestimated, lead to inadequate system design and effluent radon activities which exceed the design goal.

2. The steady state adsorption-decay constant (K_{ss}), which is a critical component of the GAC design model varied over time, was site specific, and differed by 22% to 89% from the value previously reported in the literature for the carbon tested (Barnebey-Cheney Type 1002). The factors which will most likely influence K_{ss} for a given site are flowrate and radon concentration which affect mass transfer and adsorption kinetics, and raw water quality which affects adsorption capacity. Therefore, pilot scale testing should be performed at each site to determine the appropriate K_{ss} to be used in the design model.

3. The data indicated that the GAC units retained uranium 238 and 235, and radium-226 in quantities (pCi/kg) high enough to classify the carbon as a low level radioactive waste according to State of New Hampshire regulations. The units also retained lead-210 which is not currently regulated in the State of New Hampshire. Therefore, the GAC disposal costs could increase the production cost of a GAC system. Concentration of uranium, radium and lead by GAC systems appears to be related to raw water quality (e.g., pH and alkalinity). Changes in water quality (e.g., installation of a new well) should be monitored to determine if radionuclides are released to the water supply.

4. The GAC units accumulated iron, manganese and particulates (turbidity). In addition, there were significant numbers of microorganisms growing on the carbon. As a result, GAC systems may require periodic backwashing to prevent significant headloss development. In certain water supplies, concentrations of iron, manganese and particulates may be high enough to warrant frequent backwashing which would increase operating costs and reduce the hydraulic capacity of the system. In addition, previous studies have shown that frequent backwashing may decrease radon removal efficiency. Therefore, in these cases, pretreatment for iron, manganese and particulates is recommended to decrease the backwashing frequency.

5. Gamma/beta emissions measured at the surface of the GAC units were substantially greater than background measurements. Some form of shielding will be required to lower these emissions to acceptable levels.

6. The total production costs for the GAC system at Mont Vernon, NH were $0.95/1,000 gallons without pretreatment and GAC disposal, $2.45/1,000 gallons with pretreatment and $2.64/1,000 gallons with pretreatment and disposal. (Disposal costs were based upon an estimated 20 year design life for the GAC system). During continuous monitoring, the effluent radon activity averaged 24,653 ± 17,016 (influent activity = 210,491 ± 41,384 pCi/L; flowrate = 36 ± 12 m^3/day). For Amherst, these costs were $0.56/1,000 gallons, $2.06/1,000 gallons and $2.15/1,000 gallons, respectively. The effluent radon activity of the Amherst GAC system averaged 12,000 ± 5,900 pCi/L during continuous monitoring (influent activity = 41,800 ± 15,200 pCi/L; flowrate = 61 ± 5 m^3/day). The differences in total production costs between the sites reflect the greater volume of GAC at Mont Vernon (1.33 m^3 vs. 0.85 m^3) which was a function of the higher radon loading. Since production costs for a GAC system are highly dependent on the type, volume and disposal of the carbon which are site specific parameters, extrapolation of these cost estimates to other water supplies may be inappropriate.

7. Further studies should be conducted to examine the effects of backwashing and raw water quality parameters (e.g., organics and iron) on radon removal efficiency in GAC systems. In addition, the factors affecting the retention/release of radionuclides such

as uranium, radium and the radon progeny should be investigated.

DIFFUSED BUBBLE AERATION

1. When designing a diffused bubble aeration system to remove radon from a small community water supply, it is imperative to have good data on water flowrates and influent radon activities. These data are major inputs into the design model for determining hydraulic detention time, air flowrate and diffuser configuration. As observed in this study, for small community supplies, variations in water flowrate and influent activity may be substantial and will, if underestimated, lead to inadequate system design and effluent radon activities which exceed the design goal.

2. The percent radon removal in a diffused bubble system is directly proportional to the global liquid mass transfer coefficient (K_La), the detention time and the concentration gradient ($C_{liquid} - C_{gas}$). These findings are analogous to those reported for other more conventional contaminants. Since K_La, detention time and concentration gradient are system specific, extrapolations of performance data obtained at one site should not be made to systems with other configurations/diffusers, low influent activities and/or those required to meet a more stringent MCL. Therefore, pilot-scale studies may need to be conducted at each site to obtain the required design information.

3. The total production costs for the diffused bubble system at Derry, NH were $0.64/1,000 gallons without pretreatment and $2.14/1,000 gallons with pretreatment. When operating at A:W ratios $\geq$ 5:1, at both high water flowrate (0.10 ± 0.0019 m^3/min; influent radon activity = $65,487 \pm 5,657$ pCi/L) and low water flowrate (0.047 ± 0.00053 m^3/min; influent radon activity = $78,385 \pm 6,120$ pCi/L), the overall radon removal efficiency ranged from 90.0% to >99.6%. It is possible to determine the minimum air flowrate required to meet the design removal efficiency. However, for the range of water flowrates encountered in many small communities the cost effectiveness of a larger constant speed blower (i.e., capital and operating costs) outweighs the advantages of using a smaller blower capable of delivering the minimum air flowrate. Since production costs are highly dependent

on the process equipment and percent removal required, which are site specific, extrapolation of the cost estimates for Derry to other water supplies may be inappropriate.

4. Off-gas radon activities at the Derry site ranged from 3,361 to 18,356 pCi/L in the air exiting the diffused bubble system and were 10^4 to 10^5 times higher than ambient air levels. Therefore, the impact of the off-gas on ambient air quality should be considered.

5. Though precipitation of iron and manganese was not observed during the short-term diffused bubble testing, it is well documented that this can occur in aeration systems treating groundwater, resulting in operational problems. Therefore, raw water quality should be monitored to determine whether pretreatment is required.

6. In northern climates, diffused bubble systems, including the blower intake, are typically located inside the pumphouse. Since it has been shown in this study that the radon activity in the pumphouse air may be high (as at Mont Vernon, NH) use of this air may significantly affect the ability of the diffused bubble system to meet a stringent MCL due to mass transfer limitations. Therefore, the radon activity of the influent air should be considered in the design.

PACKED TOWER AERATION

1. When designing a packed tower aeration system to remove radon from a small community water supply, it is imperative to have good data on water flowrates and influent radon activities at the site. These data are major inputs into the model for determining tower design (e.g., packing height). As observed in this study, for small community supplies, variations in water flowrate and influent activity may be substantial and will, if underestimated, lead to inadequate system design and effluent radon activities which exceed the design goal.

2. The percent radon removal in a packed tower aeration system is directly proportional to the global liquid mass transfer coefficient (K_La), the hydraulic detention time (packing height) and the concentration gradient ($C_{liquid} - C_{gas}$). These findings are analogous to those reported for other more

conventional contaminants. Since the K_La, detention time and concentration gradient are system specific, extrapolations of performance data obtained at one site should not be made to estimate percent radon removals for systems with other tower configurations/packing media, low influent activities and/or those required to meet a more stringent MCL. Therefore, pilot-scale studies should be conducted at each site to obtain the required design information.

3. For some small communities which have high influent radon activities, the packing height required to meet a stringent MCL may be greater than the 4 to 12 m range used in standard practice, possibly eliminating packed tower aeration as a viable treatment alternative.

4. The total production costs for the packed tower system at Mont Vernon, NH were $0.60/1,000 gallons without pretreatment and $2.10/1,000 gallons with pretreatment. The radon removal efficiencies ranged from 92.7% to 99.8% in spite of variation in water flowrate (0.18 to 2.6 m^3/hr), influent radon activity (115,255 to 278,488 pCi/L) and packing type (Glitsch^R mini rings and saddles, and Koch^R pall rings) over the air flowrates tested (1.75 - 51 m^3/day). Since production costs are highly dependent upon process equipment and installation, which are site specific, extrapolation of these cost estimates to other water supplies may be inappropriate.

5. The off-gas radon activities ranged from 2,410 to 21,200 pCi/L in the air exiting the packed tower and were 10^4 to 10^5 times higher than ambient air levels. Therefore, the impact of the off-gas on ambient air quality should be considered.

6. Though precipitation of iron and manganese was not observed during the short-term packed tower testing, it is well documented that this can occur in aeration systems treating groundwater, resulting in operational problems. Therefore, raw water quality should be monitored to determine whether pretreatment is required.

7. At the Mont Vernon, NH site, the intake for the blower was located inside the pumphouse, which is a conventional practice used in northern climates to prevent freezing in packed towers. However, the radon

activity in the pumphouse air approached 100 pCi/L. In cases where this design approach is used, the concentration of radon in the influent air may significantly affect the ability of the packed tower to meet a stringent MCL because of mass transfer limitations. Therefore, the radon activity of the influent air should be considered in the design.

8. Because the conventional practice of locating the blower air intake indoors may not be practical for radon removal systems required to meet a stringent MCL, an outside air intake may be required. Insulation of the tower's plumbing may be critical in northern climates to prevent freezing resulting from intermittent operation.

LOW TECHNOLOGY ALTERNATIVES

1. Radon loss during water transport in a 497 m distribution system (hydraulic retention time $\leq$ 17.9 min) was not an effective and reliable means of radon removal.

2. In studies with the pilot-scale atmospheric storage tank at Derry, NH, free fall and spray nozzle entry, and coarse bubble aeration achieved radon removals ranging from 50 to 96%. Though the data show the potential of low technology alternatives, these retrofit designs are so site specific, results cannot be extrapolated to other water supplies, especially those required to meet a stringent MCL.

3. The total production cost of retrofitting the full-scale low technology coarse bubble aeration system used at Derry, NH was $0.15/1,000 gallons. This system achieved radon removals of 80% to 88%. These costs are presented as an example of the potential savings resulting from the use of low technology alternatives. However, the cost of these alternatives is very site specific and dependent on the radon removal required. Further, at some sites the requirement of sophisticated pretreatment prior to radon removal would defeat the purpose of using low technology alternatives.

4. Based on the off-gas results obtained from the diffused bubble and packed tower aeration systems, the impacts of the off-gas from the low technology alternatives on ambient air quality should be considered.

LIQUID SCINTILLATION ANALYTICAL TECHNIQUE

1. Water samples collected using the direct syringe technique had higher mean radon activities ($\leq$ 11%) than those collected in VOA bottles. However, the widespread use of this technique may be limited due to costs, safety problems associated with distribution of syringes and the difficulty in collecting samples devoid of air bubbles. Therefore, collection in VOA bottles is likely to be the preferred sampling technique. Based on the results of this research, it is recommended that a universal hose connector be used to fill the VOA bottles.
2. Sample storage in VOA bottles was primarily affected by radioactive decay, though leakage was found to account for losses up to 20% over 21 days. Therefore, maximum storage times should be established based on the MCL, the practical quantification level, radioactive decay and leakage.
3. A toluene-based scintillation cocktail yielded significantly lower count rate than mineral oil-based and Opti Fluor O cocktails. The Opti Fluor O is less expensive ($6.50/L) than the mineral oil ($31.00/L) or toluene-based ($12.00/L) cocktails and is therefore recommended.
4. The data from the extraction experiment suggest a continuous transfer of radon from the aqueous phase to the cocktail for the radium-226 standards regardless of shaking. This finding indicates that the extraction procedure should not be used to calculate the efficiency factor. However, samples should be shaken, especially those with low activities, to insure rapid transfers of radon to the cocktail.

SECTION 3

SAMPLING AND ANALYTICAL METHODS

This section outlines the procedures for sampling and the analytical methods used in all phases of the project. Techniques specific to a given part of the research are outlined within Sections 4, 5, 6, 7, and 8. A summary of all data from the quality control checks made throughout the project is provided in Appendix A.

SAMPLING MATERIALS

Table 2 shows the sampling materials, preservation, maximum holding time, and cleaning procedures for each type of sample collected. Double deionized water was transported into the field in a 50% HCl washed plastic jug and served as a water supply for field blanks which were collected for each analysis. Field spikes were only feasible for the iron and manganese analyses. All reagents used including washing acids were reagent grade unless otherwise specified.

ANALYTICAL METHODS

Temperature, pH, Dissolved Oxygen

Samples for temperature and pH were collected in 250 mL polyethylene bottles. When dissolved oxygen (DO) was measured polarographically (i.e., during GAC Phase I), analysis was performed in the 250 mL plastic bottles. For the iodometric dissolved oxygen procedure, 300 mL glass BOD bottles were used. All bottles for the three analyses were cleaned by soaking in soapy water, rinsing three times with deionized water, soaking in 50% HCl, rinsing three times in deionized water, rinsing once with double deionized water and air drying. Materials were soaked for a minimum of 60 min. Bottles and caps for temperature and pH were rinsed three times with the sample before collection.

Temperature was obtained using ASTM approved mercury thermometers (-20 to 150°C) which were calibrated monthly against an ASTM certified mercury thermometer (-8 to 32°C). Calibration was performed by obtaining measurements in an ice bath (0°C) and at room temperature (approximately 20°C) with all thermometers. The readings from the field thermometers were plotted against the readings from the certified thermometer and used as a calibration curve. Field measurements were corrected accordingly. Sample temperatures were taken by placing a thermometer in the 250 mL bottle immediately after collection. A reading was obtained after the thermometer equilibrated (~2-3 min).

TABLE 2. SAMPLING MATERIALS PREPARATION AND PRESERVATION

Analysis	Container Volume (mL)	Container Material	Preparation (Primary Cleaning Solution)	Preservation	Hold Time
Radon	20	G*	10% Extran^R (VWR Scientific)	None	4-12 hr+ (in growth)
pH	250	P**	50% HCl	None	24 hr
Temperature	250	P**	50% HCl	None	None
Dissolved Oxygen (Polarographic)	250	P**	50% HCl	None	None
Dissolved Oxygen (Titrimetric)	300	G*	50% HCl	$MnSO_4$ Alkali-iodide H_2SO_4	12 hr
Alkalinity and Turbidity	250	P**	50% HCl	4°C (iced)	48 hr
Iron and Manganese	125	P**	50% HNO_3	HNO_3 to pH<2	6 mo
Uranium Radium	3785	P**	50% HNO_3	HNO_3 to pH<2	6 mo
Microbial Numbers	60	P**	Sterilized Autoclaved 121°C @ 15 psig	4°C (iced)	24 hr

*G = Glass.

+After extraction via shaking.

**P = Polyethylene plastic.

Initially, the pH of the water samples was measured on-site with an Orion (Cambridge, Massachusetts) Model 211 portable pH meter standardized with pH 7 and pH 4 buffers. During Phase I, the weather became very cold and the field meter did not operate correctly. As a result, samples were transported to the laboratory on ice where the pH was determined as soon as possible using an Altex^R 71 pH meter (Beckman; San Ramon, CA) with a combination pH electrode. This procedure continued throughout the project. The pH meter was calibrated on each sampling day, as outlined in Standard Methods (1985) (Method 423), using pH 7 and pH 4 buffer solutions (VWR Scientific; Boston, MA). An initial reading was then taken of one of the standards. As a check on accuracy, an aliquot of that standard was analyzed every hour or every 20 samples, whichever came first. If the standard did not read within 0.10 units of the initial pH value, the meter was recalibrated.

The pH meter was also calibrated monthly using National Bureau of Standards (NBS) pH 4 and pH 7 standards to check the commercial buffer solutions. Quality control charts for accuracy and precision were made by running three standards in the order of high (10), low (4), and intermediate (7) pH values. This order was run seven times in duplicate on three separate days and the data was used to establish upper and lower control limits (EPA, 1979). Values were reported to the nearest 0.1 pH units. If the precision or accuracy limits established by these charts were exceeded during an analytical run, the meter was recalibrated, and samples since the last acceptable check on that day were reanalyzed. Periodically, samples were split and sent to the EPA Drinking Water Research Division (Cincinnati, OH) for pH analysis. Based on these samples, the project's pH analysis was within EPA (1983) limits for acceptable accuracy and precision.

Dissolved Oxygen

Dissolved oxygen (DO) was initially determined in the field using a Yellow Springs Instrument Co. (Yellow Springs, CO) YSI Model 57 portable oxygen meter. The instrument was air calibrated on site before each use as outlined by the manufacturer. The instrument's air calibration was checked monthly against a solution containing 0 mg O_2/L prepared with Na_2SO_3 and a trace of $CoCl_2$ (Standard Methods, 1985; Method 421F). Towards the end of Phase I of the GAC evaluations (November, 1986), the weather became very cold and the meter would not work properly. Thereafter, the iodometric titration method (Standard Methods, 1985; Method 421B) was used. Samples were fixed in the field with $MnSO_4$, alkali-iodide reagent and concentrated H_2SO_4. The 300 mL bottles were filled using the sampling tubes and were allowed to overflow for approximately 2-3 min. The fixed samples were transported at 4°C to the laboratory protected from the light. The samples were titrated against a solution of sodium thiosulfate (~0.025 N). The titrant's normality was checked daily against a potassium bi-iodate primary standard. One duplicate analysis was performed on each sampling day to monitor laboratory precision.

Alkalinity and Turbidity

Samples for alkalinity and turbidity were collected in 250 mL polyethylene bottles and preserved at 4°C for transport to the laboratory. Bottles and caps were rinsed three times with the sample before collection. They were cleaned by soaking in soapy water, rinsing three times with de-ionized water, soaking in 50% HCl, rinsing three times in deionized water, rinsing once with double deionized water and air drying. Bottles and caps were soaked for a minimum of 60 min.

Alkalinity was determined potentiometrically (Standard Methods, 1985; Method 403). After the initial pH of the sample was taken, the sample was titrated against 0.02N H_2SO_4 to a pH between 4.4 and 4.6, to determine the inflection point in the curve. Alkalinity was determined from the following equation:

$$\text{Alkalinity} = \frac{A \times N \times 50{,}000}{\text{mL sample}} \qquad \text{(eq. 1)}$$

where A is the volume (mL) of standard acid used in the titration and N is the exact normality of the standard acid. The normality of the acid was determined monthly with a Na_2CO_3 primary standard. Duplicate alkalinity titrations were analyzed every fifth sample to determine precision (which was acceptable in the range ±5 mg/L as $CaCO_3$). Periodically, samples were split and sent to the EPA Drinking Water Research Division (Cincinnati, OH) for analysis. Based on these results, the project's alkalinity measurements were within EPA (1983) limits for precision and accuracy.

Turbidity was analyzed on a Hach^R Model 2100A Turbidimeter following Standard Methods 214 (1985). The instrument was calibrated prior to use on the appropriate scale with the appropriate standard using sealed secondary standards manufactured by Advanced Polymer Systems, Inc. (Redwood City, CA). EPA approved primary turbidity standards supplied by Advanced Polymer Systems were used to check the secondary standards monthly. The meter was recalibrated at least every hour or every 20 samples, whichever came first. Duplicate analyses were performed every fifth sample to check precision. Precision quality control charts were constructed using the data from the first few months of the project (EPA, 1979). The precision for all of the duplicate sample analyses were combined and upper and lower control limits were calculated. Accuracy quality control charts were constructed in the same manner as the precision charts, only using the data from readback standards as the input. Each run, standards were checked and the accuracy was plotted on the accuracy control chart. If any accuracy or precision checks fell outside of the control limits, all the samples on a given day up to the last acceptable quality control check were reanalyzed. Values were reported to the nearest 0.05 NTU. Turbidity samples from the EPA (EMSL; Cincinnati, OH) water supply performance evaluation studies were analyzed in the laboratory during 1987 and 1988 as part of the quality assurance program for the project. Periodically, field samples were split and sent to the EPA Drinking Water Research Division (Cincinnati, OH) for analysis. Based on both the WS series and split sample results, the turbidity analyses for the project were within EPA (1983) limits for accuracy and precision.

Iron and Manganese

Iron and manganese samples were collected in 125 mL polyethylene bottles. The sampling materials were cleaned by soaking in soapy water, rinsing three times with deionized water, soaking in 50% HNO_3, rinsing three times with deionized water, rinsing once with double deionized water and air drying. The bottles and caps were soaked for a minimum of 60 min. Bottles and caps were rinsed three times with the sample before collection. The samples were acidified to pH<2 with concentrated HNO_3.

Iron and manganese samples were analyzed on a Perkin-Elmer® (Norwalk, CT) Model 2380 flame atomic adsorption spectrophotometer (AAS) using an air-acetylene flame and a 10 cm burner head according to Standard Methods 303A (1985). Standards were prepared daily from stock solutions of 1,000 ppm iron and 1,000 ppm manganese (EM Science; Cherry Hill, NJ). Although matrix problems were not anticipated, a $CaCO_3$ solution, outlined in Method 303A, was added to the samples and standards to combat ionization interference. Two standards, S1 and S2, for each metal were used to calibrate the AAS as described by the manufacturer. A standard curve was established for each metal using absorbances of 4 or 5 standards to monitor the linearity of the instrument based on S1 and S2 inputs. Readback standards were run after every 10 samples to check accuracy. Duplicates were analyzed every fifth sample. Concentration measurements were read directly off the AAS. Control charts were made for precision and accuracy. On each of three days, four standards of each metal were prepared and analyzed seven times as described in EPA (1979). Upper and lower control limits were calculated for accuracy using the percent recoveries and for precision using the difference between duplicates (EPA, 1979). The standards were also run at the end of each analytical run. Two samples at every site during every sampling period were randomly chosen and spiked with 25 µl each of 1,000 ppm iron and manganese. An aliquot of double deionized water brought into the field was also spiked with the same amount of the two metal stocks. Samples were periodically split and sent to the EPA Drinking Water Research Division (Cincinnati, OH) for analysis. Based on these results, the iron and manganese measurements were within EPA limits for precision and accuracy (EPA, 1983).

The GAC samples collected for metals' analysis during the coring experiment were dried to a constant weight in an oven at 105°C. Two subsamples approximately 2 g each were placed in tared 125 mL polyethylene bottles (cleaned as described above) and weighed on a Mettler AC-100 analytical balance. 50 mL of 10% HNO_3 were added to each bottle. The samples were digested according to the method outlined by Lessard (1987) for 24 hr in a 60°C water bath. The liquid was decanted, filtered through double deionized water washed GF/C filters and diluted to 100 mL in a volumetric flask. They were analyzed using the AAS according to the Standard Methods 303A (outlined above).

Microbial Numbers

Water samples to determine microbial numbers were collected in 60 mL sterile polyethylene bottles and preserved at 4°C during transport to the laboratory. The bottles were cleaned by soaking in soapy water, rinsing three times in deionized water and autoclaving at 120°C and 15 psig for 20 min. The bottles and caps were rinsed three times with the sample before collection.

Microbial populations were determined using the spread plate method. A 0.1 mL aliquot of sample was plated on tryptone glucose extract (standard plate count - SPC) agar (Difco, Detroit, MI) and incubated at 20°C for 7 days. The incubation period and temperature were determined in preliminary experiments with the water samples to yield the maximum number of organisms. The number of colony forming units (CFU) per plate between 20 and 200 were recorded using a Quebec colony counter. Aseptic techniques outlined in Standard Methods (1985, Method 907 and 907B) were followed. All samples were plated in duplicate. Normally, dilutions of 10^{-1} and 10^{-2} were adequate to achieve the correct number of colonies per plate. Sterile controls were run with every set of samples to check aseptic technique.

GAC samples for microbial analysis were collected in sterile polyethylene bags and stored at 4°C during transportation to the laboratory (~4 hr). Aliquots of approximately 5 g were placed in sterile, tared polyethylene bottles (125 mL) and weighed on a Mettler AC-100 balance. They were then mixed with 45 mL of 0.1% sodium pyrophosphate and sonicated for 30 min. Serial dilutions (10^{-1} to 10^{-8}) of the supernatant were plated in triplicate on SPC agar and subsequently counted as outlined above.

Uranium, Radium, and Lead Radionuclides

Aqueous samples for uranium and radium analyses were collected in 3.79 L polyethylene containers and acidified to pH <2 with concentrated HNO_3. Bottles and caps were rinsed three times before collection of the sample. The sampling materials were cleaned by soaking in soapy water, rinsing three times with deionized water, soaking in 50% HNO_3, rinsing three times with deionized water, rinsing once with double deionized water and air drying. The bottles and caps were soaked for a minimum of 60 min.

Samples were analyzed for total uranium and radium-226 by the State of New Hampshire Department of Environmental Services Laboratory (Concord, NH). Total uranium alpha particle activity was analyzed according to Method 908.0 (EPA, 1980) where the uranium species were coprecipitated with ferric hydroxide, redissolved in HCl and concentrated on an anion exchange column. Uranium eluant from the column was dried and counted for alpha particle activity using a Ludlum (Sweetwater, TX) 1000 scaler and a Ludlum 43-10 detector. Radium-226 was measured using the radon emanation technique (EPA, 1980). The radium was coprecipitated on barium sulfate, dissolved in EDTA and stored in a sealed chamber to allow ingrowth of radon-222. The radon gas was collected in a Lucas scintillation cell and counted using a Ludlum 1000 scaler and a Ludlum M-182 detector. A tracer spike of barium-133 was added to all radium samples.

Uranium 238 and 235, radium-226 and lead-210 activities of the GAC were determined from the coring experiment samples. After collection of approximately 200 mL of GAC per sample in 50% HNO_3 washed 227 g glass Mason jars, the materials were taken to the New Hampshire Division of Public Health Services Radiological Health Program's laboratory (Concord, NH). The contents of jars were dried to a constant weight at 105°C. The samples were then placed in the 105°C oven with a Mason jar lid on them and reheated. Immediately after removal from the oven, the screw cap lids were tightened and upon cooling the lids sealed as a vacuum was formed. This procedure was used to retain any radon subsequently produced. Samples were held at room temperature in the laboratory for 44 days before analysis.

Gamma spectroscopic analyses were conducted using a Nuclear Data (Schaumburg, IL) Model 6620 spectrometer fitted with a 13% Ge(Li) detector (10 hr counts/sample). The uranium-238 values represented the means of the activities of thorium-234 and protactinium-234, if both were found. The values of radium-226 were the means of the lead-214 and bismuth-214 activities. The GAC was assumed to have a bulk density of 428 kg/m^3 as noted by the manufacturer (Barnebey Cheney; Columbus, OH).

The samples were held in the laboratory for 1.5 years and subsequently transported to the University of New Hampshire. They were redried and counted on a Canberra high resolution gamma spectrometer with a coaxial Ge diode well detector (Model GCW 1022-7500). The system was coupled to a computer-based multi-channel analyzer (Model 35). A virgin GAC sample was counted for 15.4 hr. The GAC samples from the coring experiment were counted for 2-3.4 hr depending on their activities. Data was reduced according to the method outlined by Larson and Cutshall (1981) to determine the activity/kg of uranium 238 and 235, radium-226 and lead-210.

Gamma/Beta Emissions

Gamma/beta measurements were obtained by placing the unshielded detector of a Victoreen (Cleveland, OH) THYAC IV survey meter (Model 290) on the surface of the treatment units or holding it exposed to the air. A Geiger-Müeller probe (Model 491-40) was used and the meter set to record mR (milliRoentgen) dose rate per hr^{-1}. The meter was calibrated against a cesium-137 source as outlined by the manufacturer (Victoreen, 1985).

Radon

Aqueous samples for radon were drawn directly from the plastic sampling tubes or from inverted funnels filled overflowing with water using 10 mL Hamilton^R gastight glass syringes equipped with 13 gauge needles. The 13 gauge needles were used to minimize cavitation when the sample was drawn. The syringe was rinsed three times with the sample before collection. The 10 mL sample, devoid of air bubbles, was slowly injected into a 20 mL capacity low-absorbance liquid scintillation vial below the surface of 10 mL of scintillation cocktail [40.3 mL Liquifluor^R (Dupont, Wilmington, DE) per 1 L of scintillation grade toluene] already in the vial. The vials and syringes were cleaned before each use by soaking in soapy water, rinsing three times in deionized water, soaking in Extran 1000^R (VWR Scientific, Boston, MA), rinsing four times with deionized water and air drying. All materials were soaked for a minimum of 20 min.

The samples were analyzed once radon came to secular equilibrium, with its short-lived progeny (preliminary experiments indicated we should wait 4 hrs after collection) and before 12 hrs had elapsed between collection and analysis. Radon samples were analyzed using a Beckman (Fullerton, CA) LS 7000 liquid scintillation counter (LSC). Program 6 was used for all analysis (Appendix B). The counting time was 10 min per sample. Automatic quench compensation was used with a cesium-137 source. The percent error in channels 1 and 2 was two times the standard deviation. The instrument was calibrated with Beckman tritium, carbon-14 and background sealed standards. A radium-226 standard containing 260.85 disintegrations per minute (dpm) (approximately = 117.5 pCi/L) was used for daily standardization. The counts per minute (cpm) value of this standard, which contained a known amount of radon-222 in secular equilibrium with radium-226, was used to calculate a counting efficiency (E) for the instrument. The LSC provided results in counts per minute (cpm) for two windows: Channel 1 = 0 - 397 (corresponding to a full tritium channel) and a Channel 2 = 397 - 940 (a tritium - phosphorus-32 channel). The majority of the counts from radon-222 and its progeny were located in the channel 2 pulse heights. The total counts (Channel 1 and 2) for each sample were then converted to pCi/L using the equation:

$$A = \frac{(C_s - C_b) \times 1000 \text{ ml/L}}{(E)(V)(D)} \qquad \text{(eq. 2)}$$

where:
A = radon activity (pCi/L)
C_s = cpm of the sample
C_b = background cpm of field blank (cpm)
E = efficiency factor (cpm/pCi)
D = decay correction factor
V = sample volume (mL)

The decay correction was determined from:

$$D = e^{-0.693(T)/t^{1/2}}$$

where:
T = time from sampling to counting (days)
$t_{1/2}$ = half-life of radon (3.82 days)

Duplicate samples were analyzed every tenth sample to determine precision. The radium-226 standard and field blank were counted at the beginning of each analytical run.

The radium-226 standards were prepared from NBS or EPA (Environmental Monitoring Systems Laboratory; Las Vegas, NV) primary standards. The standards, in glass ampules, typically contained approximately 5 g of an HCl solution with radium-226 activities ranging from 4 to 5 nCi/g. The contents of the ampule were emptied into a 50% HNO_3 washed and air dried glass beaker to facilitate weighing in a Mettler AC-100 analytical balance. The contents of the beaker were then transferred to a 200 mL volumetric flask (cleaned using 10% Extran^R, 50% HNO_3 and double deionized water) and the beaker was rinsed several times with 0.05M HNO_3 to insure all radium-226 was transferred. The stock radium-226 solution was prepared in a balance of 0.05M HNO_3. It was stored in the wax-sealed 200 mL volumetric flask at 4°C. The stock solution usually contained 117.5 pCi/mL (260.85 dpm/mL) of radium-226. 5 mL of the stock solution was added using a volumetric pipette to a clean scintillation vial and diluted with 5 mL of 0.05M HNO_3. The contents of the vial were purged for 20 min with fine bubbles of laboratory grade nitrogen gas. Subsequently, 10 mL of the toluene-based scintillation cocktail were slowly added to the standard. The scintillation vial was sealed and counted daily on the LS 7000 to determine when the radium-226 was in secular equilibrium with radon-222 (~20-25 days). In most cases, standards were used for a period of 2-3 months before replacement. Once the standard was in secular equilibrium it was used. If the cpm of the radium standards began to change before that time, they were no longer used. Samples were sent regularly to the EPA Environmental Monitoring and Support Laboratory (Cincinnati, OH) for interlaboratory checks until it closed. Duplicates were analyzed every 10 samples to determine precision. The detection limit for the UNH radon analysis was determined according to the procedures outlined in EPA (1984) to be 139 pCi/L for a 10 min counting time.

The activity of radon in the off-gas was monitored for the diffused bubble and packed tower aeration systems using a Pylon^R AB-5 portable radiation monitor (Ontario, Canada). The monitor was programmed to pump air (0.5 L/min) for 10 min through a hose into a Model 300 Lucas Cell. After 10 min, the cell was sealed and the radon allowed to come to secular equilibrium with its progeny (~3 hrs). The radiation monitor came equipped with a 3368 dpm radium-226 standard calibration cell. The efficiency of the machine was determined daily using this standard. The efficiency was used to convert sample cpm to pCi/L. A machine efficiency of approximately 73% was maintained according to the manufacturer's specifications. The procedures used were outlined in the manufacturer's guide (Pylon, 1987).

Efficiencies (E) were calculated in percent by:

$$E = \frac{\text{standard cell cpm}}{\text{standard cell dpm}} \times 100\% \qquad \text{(eq. 3)}$$

Sample radon activities were found using the average stable cpm after approximately 3 hr of ingrowth in the equation:

$$pCi/L = \left[\frac{C_s - C_b}{3 \times 2.22 \times 0.285 \times E}\right] \times 100 \qquad \text{(eq. 4)}$$

where:

C_s = cpm found for sample
C_b = cell background cpm
3 = number of alpha particles emitted during radon's decay
2.22 = conversion factor (dpm/pCi)
0.285 = volume of Model 300 sample cell (L)

The monitor had a maximum count rate of 10^6 per second.

Percent Moisture Content

GAC samples were collected during the coring experiment and placed in sealable polyethylene bags. They were transported to the laboratory where approximately 30-40 g aliquots were placed in tared aluminum dishes. They were subsequently weighed on a Mettler AC-100 analytical balance and placed in an oven at 105°C until a constant weight was attained. The samples were cooled in a dessicator and then reweighed on the balance. The change in weight was used to calculate the percent moisture content of the samples. These data were used to correct all coring analyses to kg of GAC (dry weight).

SECTION 4

GRANULAR ACTIVATED CARBON

INTRODUCTION

The ability of granular activated carbon (GAC) to remove radon has been demonstrated by Lowry (1985) who reported removals of 80-99.9% for waters containing 40,000-750,000 pCi/L using point-of-entry systems with 0.028 to 0.071 m^3 of GAC. However, less information is available on the efficacy of using GAC for radon removal in small community systems (Lowry and Lowry, 1988; Reed, 1989). In this phase of the research, downflow GAC systems installed at two mobile home parks located in Amherst and Mont Vernon, NH were studied. The Mont Vernon GAC system had been operating for approximately 1 yr before the study began. For this research project, the existing carbon was removed and replaced with virgin GAC.

The water system at Amherst serves 56 homes at an average daily flow of 59 ± 4 m^3/day. The water is obtained from one well containing an average radon activity of 49,500 ± 11,200 pCi/L. The water system at Mont Vernon supplies 40 homes at an average flow of 31 ± 11 m^3/day. Water is obtained from two wells with an average radon activity of 222,000 ± 52,000 pCi/L. The well water in both systems was pumped to atmospheric storage tanks and subsequently, pumped through the GAC systems upon demand from the community. A schematic of both systems is provided in Figures 1 and 2.

The systems were evaluated based on radon removal efficiency, retention of radionuclides, gamma/beta emissions, and economics. Radon and several general water quality parameters (alkalinity, turbidity, dissolved oxygen, temperature, pH, iron, manganese and bacterial numbers) were monitored at each site. Uranium and radium were also monitored in the water supply. The GAC was cored and analyzed for accumulation of uranium-238, uranium-235, radium-226, lead-210, iron, manganese and microbial numbers. Gamma radiation measurements were taken at the surface of the units, and at locations inside and outside of the pumphouses to determine whether exposure presented significant health and safety problems.

An economic analysis of the GAC systems was performed to determine the costs of using GAC for radon removal from small community drinking water supplies. The analysis included capital, operation and maintenance, and disposal costs. In addition, the total annual cost and the production cost for each GAC system was estimated.

There are several advantages of using GAC systems for radon removal in small community water supplies: (i) GAC units can be installed in-line so no additional pumping is required (systems are already under pressure), (ii)

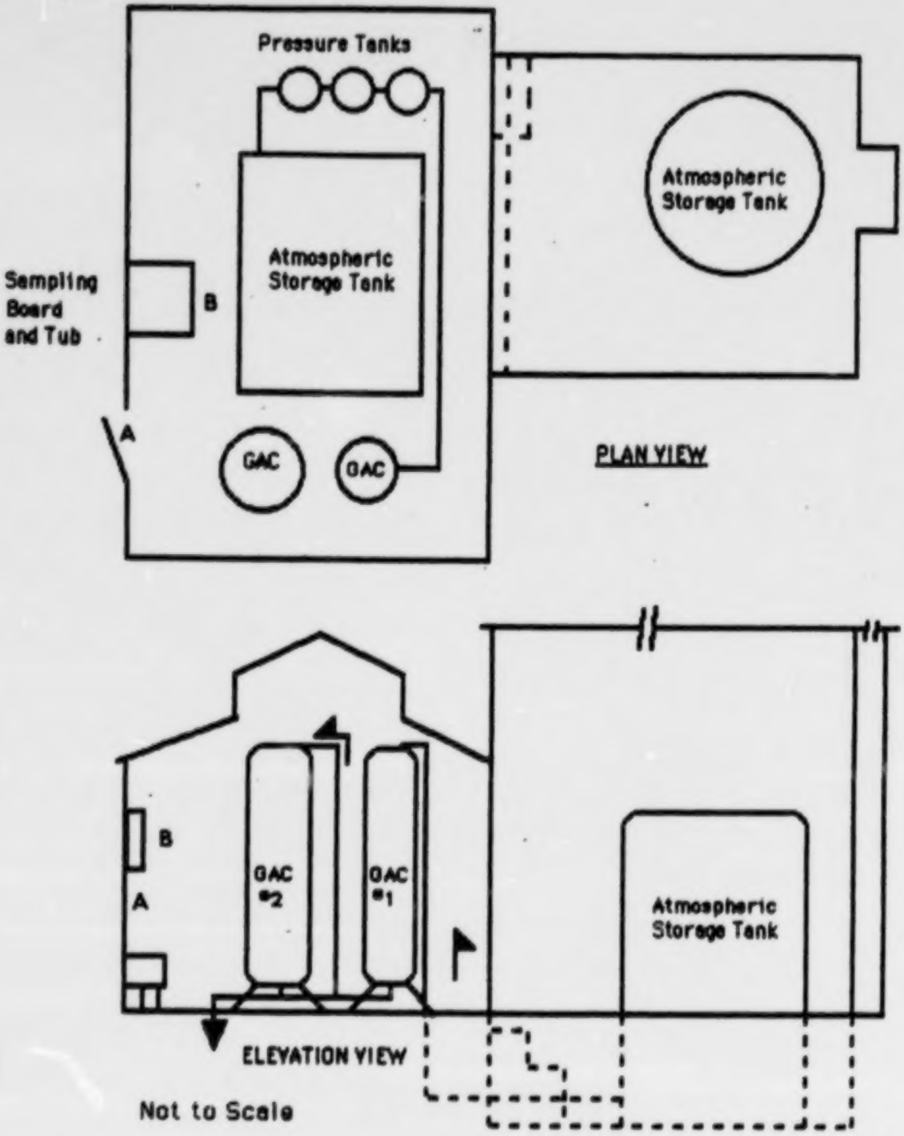
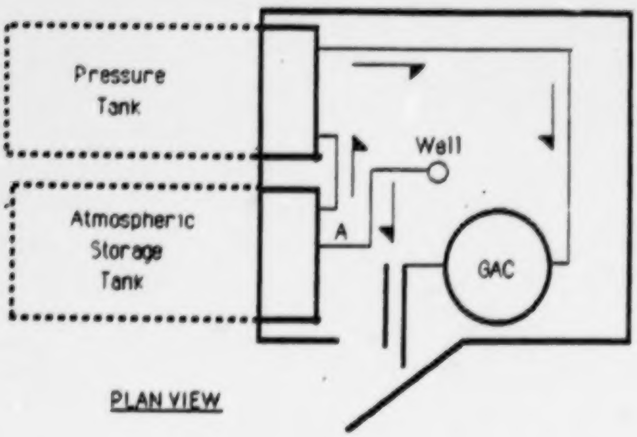
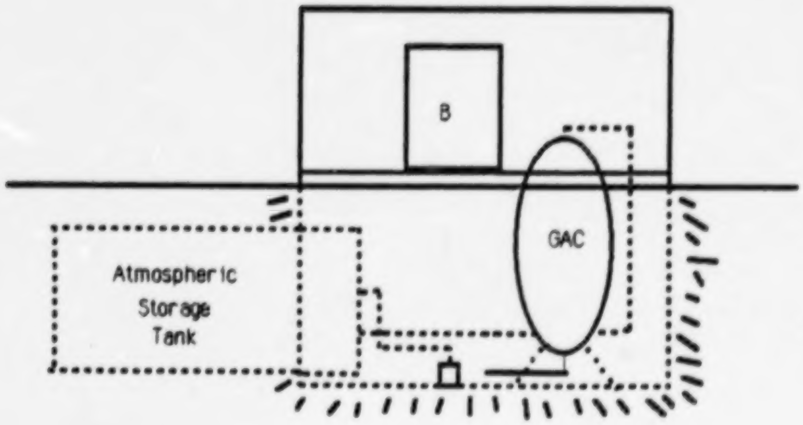

Pressure Tanks
Atmospheric Storage Tank
Sampling Board and Tub
B
Atmospheric Storage Tank
A
GAC
GAC
PLAN VIEW
B
A
GAC #2
GAC #1
Atmospheric Storage Tank
ELEVATION VIEW
Not to Scale

Figure 1. Schematic of the Mont Vernon, NH water supply and GAC system (not to scale). A and B are locations of gamma/beta emission measurements.


Pressure
Tank
Well
Atmospheric
Storage
Tank
A
GAC

PLAN VIEW


B
GAC
Atmospheric
Storage
Tank

ELEVATION VIEW

Not to Scale

Figure 2. Schematic of the Amherst, NH water supply and GAC system (not to scale). A and B locations for gamma/beta emission measurement.

radon is contained, not released into the air, and (iii) GAC has lower maintenance requirements than aeration systems and has no moving parts. The disadvantages of GAC are: (i) the potential for substantial gamma emissions from radon progeny, (ii) the potential for accumulation of long-lived radionuclides (i.e., uranium 238 and 235, radium-226 and lead-210), (iii) the long empty bed contact time (EBCT) required, and (iv) the potential for fouling of the GAC by oxidized metals, organics, particulates, and/or microorganisms, requiring frequent backwashing.

EXPERIMENTAL PROCEDURES

Description of Units

The GAC systems units were designed by Dr. Jerry Lowry (Unity, ME) and started on October 8, 1986. The GAC used for the research project, Barnebey Cheney 1002, was chosen based on previous studies by Lowry (1985) and placed in the fiberglass tanks installed in the pumphouses. After the GAC was in place, the units were backwashed to remove fines. At Amherst, the system consisted of one 91.44 cm diameter contactor containing 0.85 m^3 of GAC. At Mont Vernon, the system consisted of two contactors in series; a 76.2 cm diameter contactor (GAC #1) containing 0.57 m^3 of GAC followed by a 91.44 cm diameter contactor (GAC #2) containing 0.76 m^3 of GAC for a total of 1.33 m^3 of GAC. The system at Mont Vernon was designed with two units because of the high influent radon activity.

Sampling taps for water samples were installed on the influent and effluent lines and at intervals within each unit. The sampling taps before and after the units were constructed of Schedule 40 PVC tees and brass valves. Taps within each unit were constructed of 1.91 cm diameter Schedule 80 PVC pipe. The PVC pipe was slotted laterally with 0.0305 cm slots and extended 22.86 cm into the GAC bed. The pipe was supported by a 1.91 cm PVC bulkhead installed in the contactor wall. Six millimeter plastic tubing was connected to the bulkhead which extended to a sampling board containing brass valves. The sampling tap locations for the Amherst and Mont Vernon systems are shown in Figures 3 and 4, respectively. The sampling tap locations corresponded to the GAC volumes shown in Table 3.

Experimental Design

The sampling frequency was varied to meet the needs of each experimental phase. The systems were monitored daily for 3 to 4 days and then every 2 to 5 days for approximately one month during the initial operating period (Phase I). Thereafter, during Phase II, they were monitored weekly, biweekly and then monthly. The system at Amherst was monitored for 122 days, while the system at Mont Vernon was monitored for 478 days. Several special sampling events were performed on the GAC systems, including coring the GAC material, diurnal observations, exposure to high flow and backwashing.

Coring

All GAC units were cored on July 29, 1987. Solid GAC samples were obtained by forcing a coring apparatus through the material as the bed was

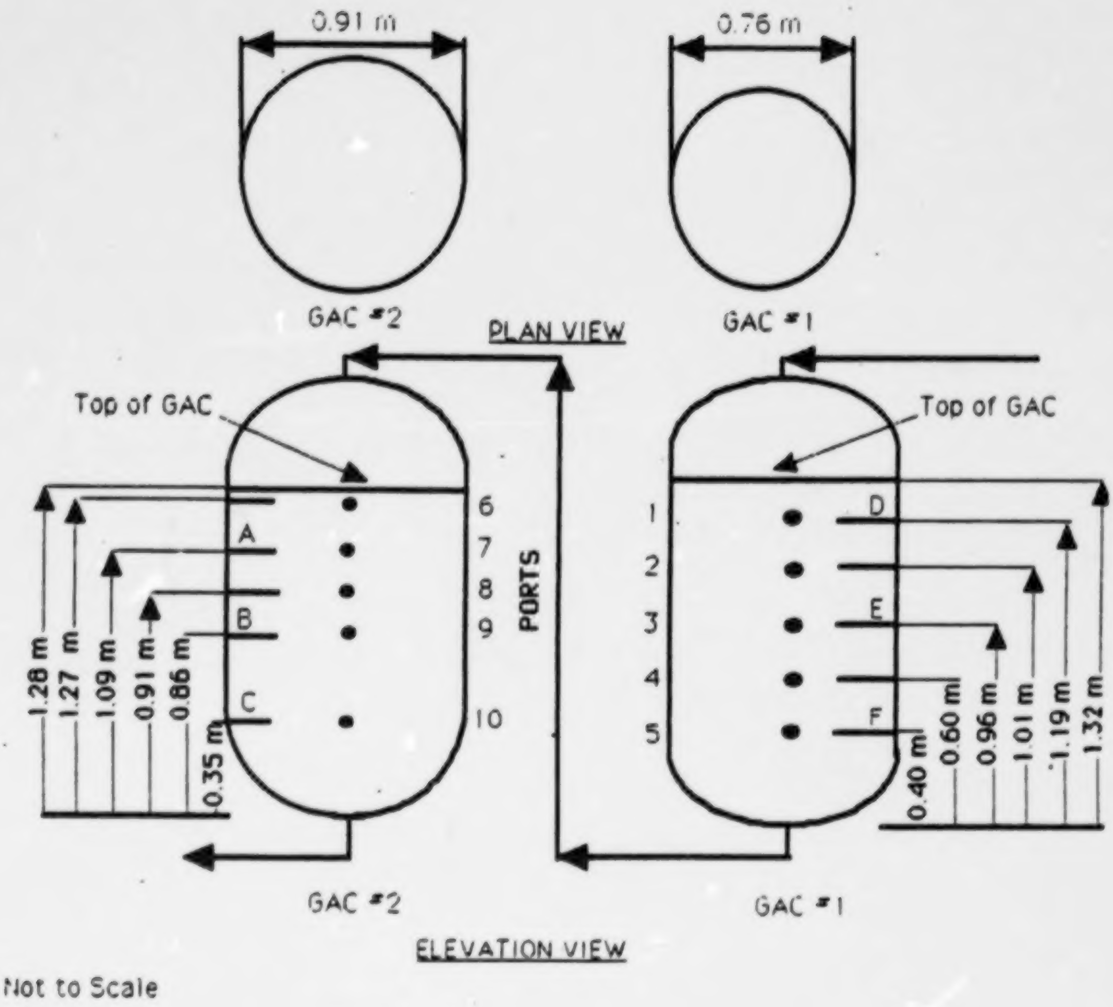

0.91 m
0.76 m
GAC #2
PLAN VIEW
GAC #1
Top of GAC
Top of GAC
6
7
8
9
10
A
B
C
PORTS
1
2
3
4
5
D
E
F
1.28 m
1.27 m
1.09 m
0.91 m
0.86 m
0.35 m
0.40 m
0.60 m
0.96 m
1.01 m
1.19 m
1.32 m
GAC #2
GAC #1
ELEVATION VIEW
Not to Scale

Figure 3. Schematic of the Mont Vernon, NH GAC system. A,B,C,D,E, and F are locations of gamma/beta emissions measurements.

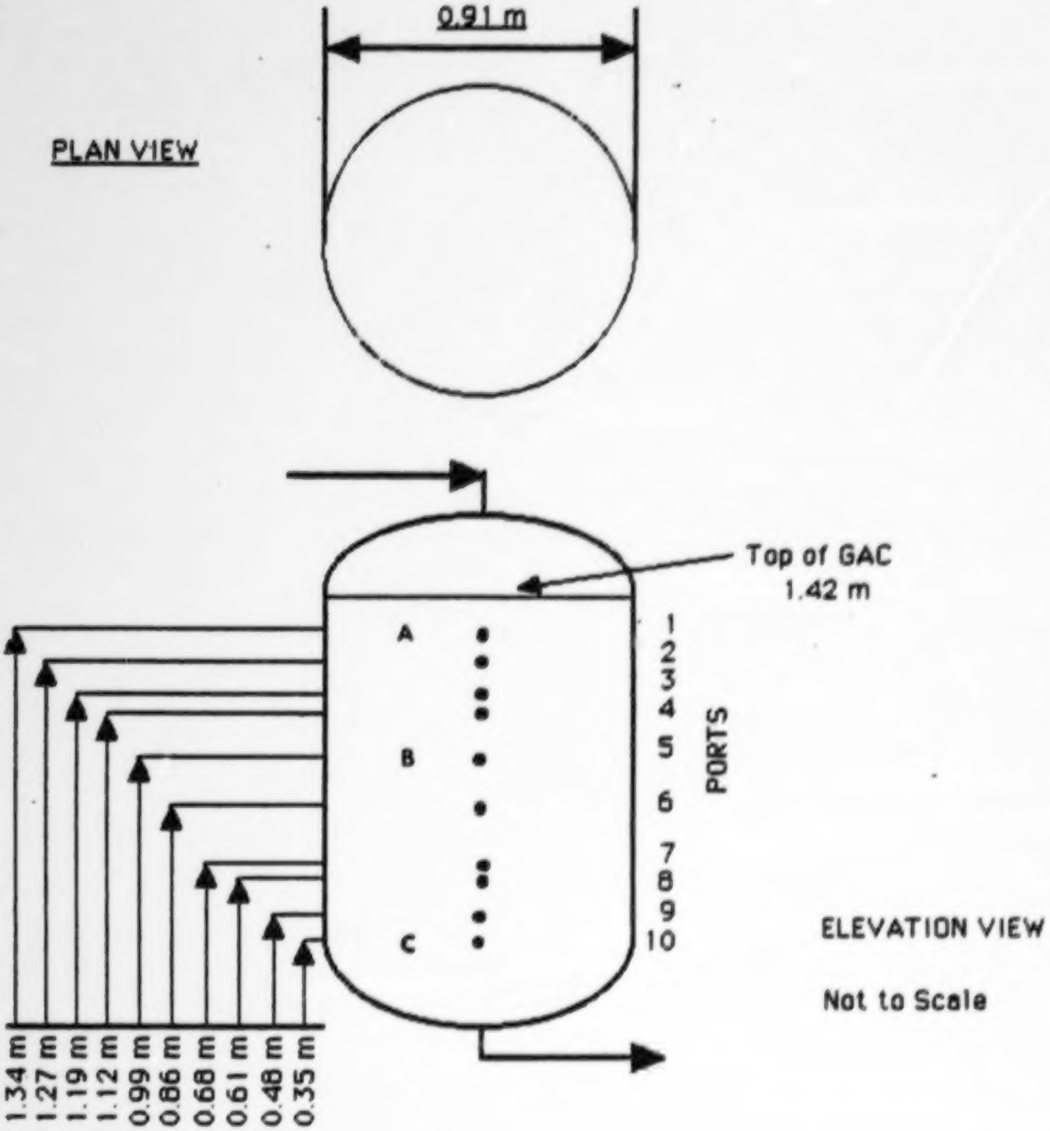

0.91 m
PLAN VIEW
Top of GAC
1.42 m
A
B
C
1
2
3
4
5
6
7
8
9
10
PORTS
ELEVATION VIEW
Not to Scale
1.34 m
1.27 m
1.19 m
1.12 m
0.99 m
0.86 m
0.68 m
0.61 m
0.48 m
0.35 m

Figure 4. Schematic of the Amherst, NH GAC system. A, B and C are locations of gamma/beta emission measurements.

TABLE 3. RELATIONSHIP BETWEEN SAMPLING PORTS AND GAC VOLUMES

Port Number	Mont Vernon GAC Volume Between Ports (m³)	Mont Vernon Total GAC Volume (m³)	Amherst GAC Volume Between Ports (m³)	Amherst Total GAC Volume (m³)
Influent				
1	0.058	0.058	0.050	0.050
2	0.081	0.14	0.050	0.10
3	0.081	0.22	0.050	0.15
4	0.081	0.30	0.050	0.20
5	0.12	0.42	0.084	0.28
6	0.16	0.58	0.084	0.37
7	0.12	0.69	0.084	0.45
8	0.12	0.81	0.084	0.53
9	0.12	0.93	0.084	0.62
10	0.25	1.17	0.084	0.70
Effluent	0.16	1.33	0.150	0.85

slightly expanded with backwash water (raw water from the storage tank). The coring device was a 2.54 cm diameter (I.D.) plexiglass tube 4 cm O.D. with 7.62 cm, sampling compartments centered every 10.16 cm. Samples of the bottom 36 cm, middle 36 cm and remaining material on top were composited. The samples were placed in sterile polyethylene bags and half-pint glass Mason jars.

The GAC samples in the jars were analyzed for uranium-238, uranium-235, and radium-226 by the State of the New Hampshire Radiological Laboratory and the Institute of Earth, Oceans, and Space at the University of New Hampshire (UNH). The UNH facility also analyzed the samples for lead-210. The GAC samples were also analyzed for percent moisture content, microbial numbers, iron and manganese.

Diurnal Variations

The units at Mont Vernon were sampled on July 1 and 2, 1987 to investigate the effect of diurnal variations in the water supply on the performance of the system. Samples were collected from the influent and effluent ports every hour for 12 hr (13 sampling times). The samples were analyzed for all parameters except microbial numbers, gamma/beta emissions, uranium and radium.

High Flow

On July 16, 1987, the units at Mont Vernon were sampled to observe the effects of high flow on the performance of the system. A tap installed after GAC #2 allowed the sampling team to waste 33 m^3/day of water in addition to the community demand, creating an artificially high flow through the GAC system. This represented a 91% increase in demand above the average flowrate. Samples were collected over a 3 hr period until the wells could not sustain the demand. Radon samples were collected from all ports. Samples for all other analyses were collected from the influent and effluent ports only. Uranium and radium analyses were not performed during this event.

Backwashing

The units at Mont Vernon were backwashed on January 28, 1988. The system was designed to be backwashed at 12.2 m^3/m^2-hr. This flowrate was too high, creating excessive loss of GAC. The backwash rate was lowered immediately to 55 m^3/day or 5.0 m^3/m^2-hr for GAC #1 and 3.4 m^3/m^2-hr for GAC #2. The units were backwashed separately for 10 min using raw well water from the storage tank.

Samples were collected prior to backwashing, as well as 1.3, 2.6, 3.9 and 24 hr after backwashing. Radon samples were collected from all ports. Samples for all other analyses, except uranium and radium, were collected from the influent, effluent and ports 1, 3, 5, 6, 8, and 10. Samples of the backwash water effluent were also collected at time = 0 and time = 10 min. These samples were analyzed for radon, iron, manganese, uranium and radium.

Sampling and Analysis

Water samples were collected directly from the plastic tubes connected to the sampling ports and flow was controlled using brass valves. The tubes were routed to a sampling board located some distance from the units to minimize the gamma radiation exposure to sampling personnel. Tubing was allowed to flush for 3 min before sampling commenced. Gamma/beta emissions were measured on the outside surface of the GAC contactors using a Geiger Müeller (GM) survey meter placed at locations shown in Table 4 and Figures 1 and 2. Flow was monitored using a flow totalizer downstream of each GAC system.

RESULTS AND DISCUSSION

Phase I - Initial Operation

Water samples were collected from the system at Mont Vernon daily for 4 consecutive days and every 2 to 5 days for approximately 32 days thereafter. Water samples were collected from the Amherst GAC system daily for 3 consecutive days and every 2 to 5 days thereafter. On October 12, 1986 (Day 4), the system at Amherst was shutdown because of problems with the high and low flow electrodes in the atmospheric storage tank. The system was started again on October 16.

By the end of Phase I, sufficient data on each system's operating characteristics were collected to allow sample frequency to be decreased to once per week.

pH and Alkalinity--

Mont Vernon site--The pH and alkalinity of the water changed dramatically through the GAC units during the first days of operation. The influent pH remained constant at 6.5 ± 0.20, while the effluent pH decreased from 9.9 to 6.5. There was a substantial increase in pH from Day 1 to Day 15 as the water passed through the units (Figure 5). The effluent pH remained significantly higher ($\alpha = 0.05$ and 0.01, Student's t test [StT]) than the influent throughout Phase I.

A similar increase through the unit was observed for alkalinity from Day 1 to 29 (Figure 5). The alkalinity increased markedly as the water passed through the units during the first days of operation. During the latter part of Phase I, the increase in alkalinity through the GAC system was less substantial, but still statistically significant ($\alpha = 0.05$ and 0.01, StT).

Amherst site--As with the Mont Vernon system, the pH of the water in Amherst was affected during Phase I. The influent pH averaged 7.9 ± 0.26, while the effluent pH decreased from 9.5 to 7.8. Figure 6 shows the marked increase in pH as the water passed through the unit from Day 1 until Day 13. During the remainder of Phase I, there was no significant difference between influent and effluent pH ($\alpha = 0.05$ and 0.10, StT). The influent pH of the Amherst water was significantly ($\alpha = 0.05$ and 0.01, StT) higher than the Mont Vernon water's pH (6.5 ± 0.20).

TABLE 4. LOCATIONS OF GAMMA/BETA EMISSIONS MEASUREMENTS* ON GAC UNITS

Mont Vernon		Amherst	
Port Number	Total+ GAC Volume (m^3)	Port Number	Total+ GAC Volume (m^3)
1	0.058	1	0.05
3	0.22	5	0.28
5	0.42	10	0.70
6	0.58		
8	0.81		
10	1.17		

*Measurements were made on the outside surface of the GAC contactors.

+Based on distance from top of GAC in contactors .

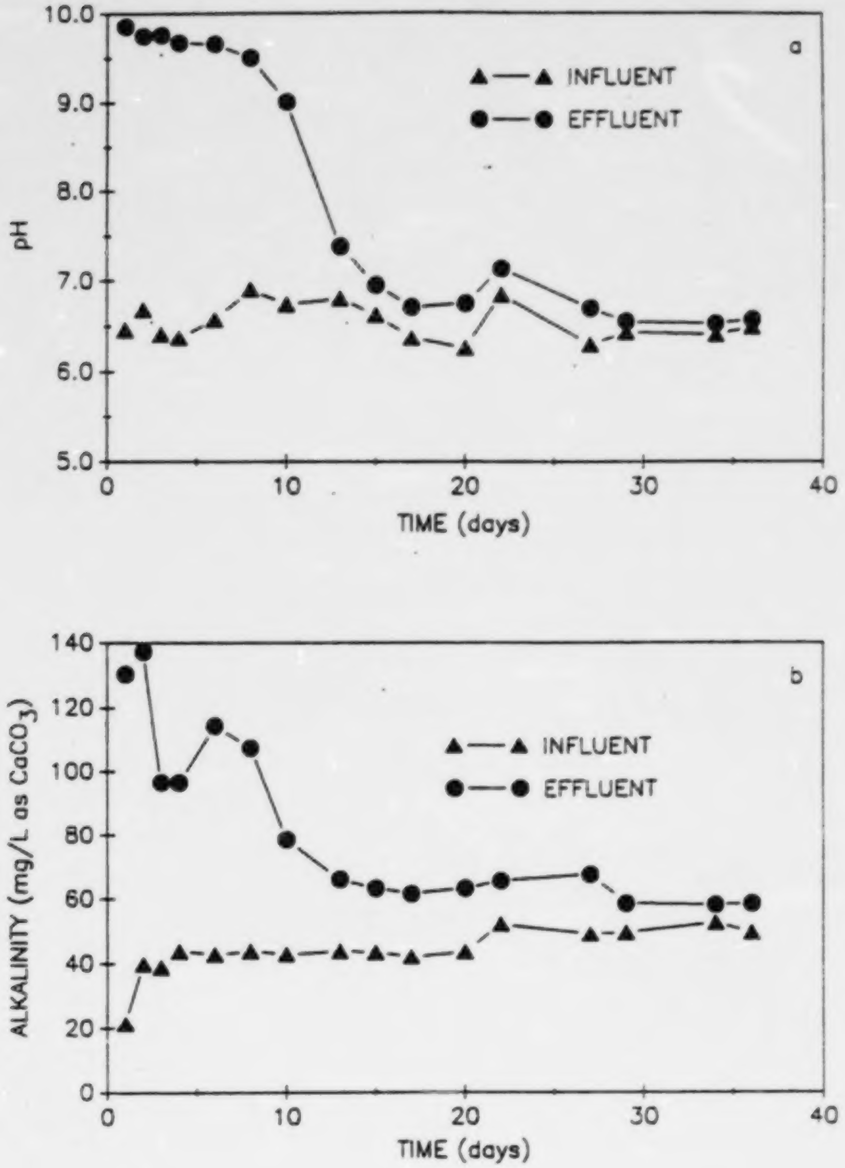


Figure 5. Phase I - Mont Vernon, NH GAC system. pH (a) and alkalinity (b) data for first 36 days operation.

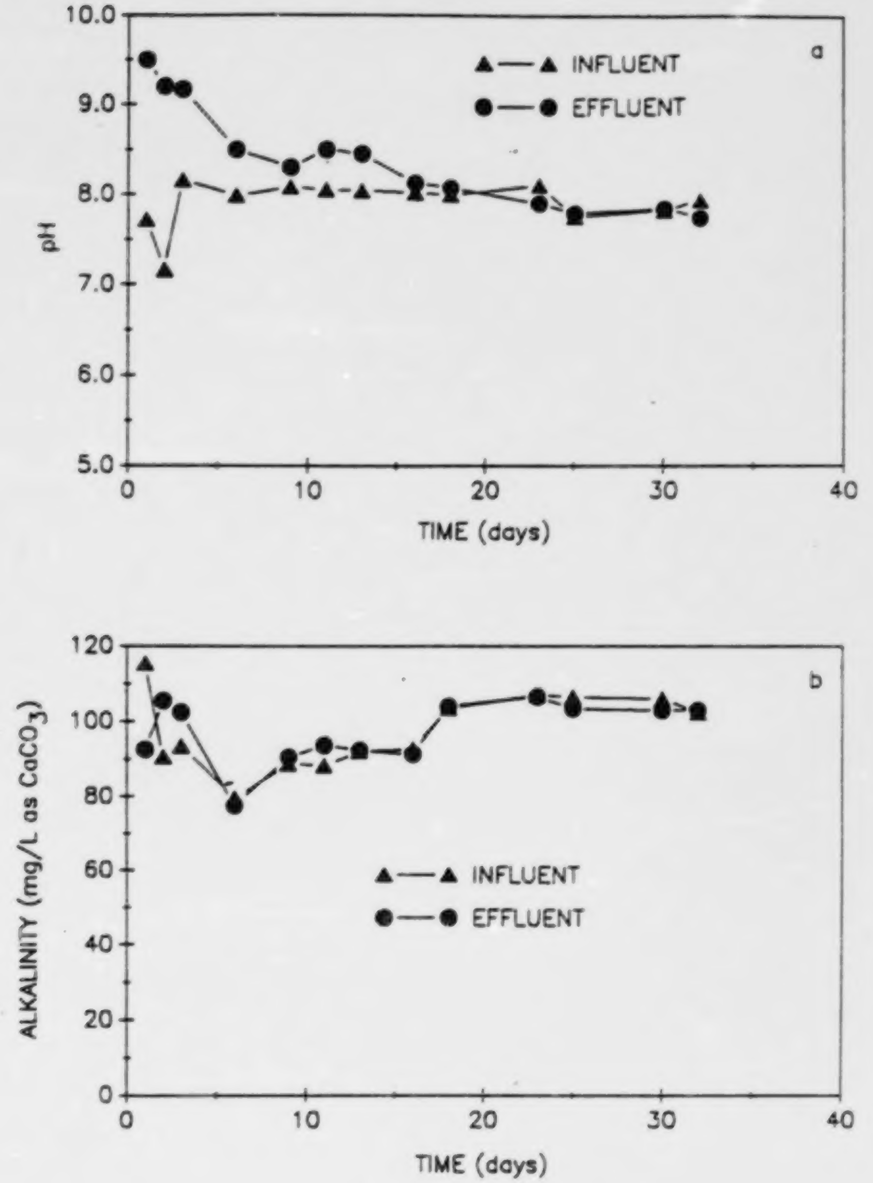


Figure 6. Phase I - Amherst, NH GAC system. pH (a) and alkalinity (b) data for first 32 days of operation.

The influent (97 ± 10.1 mg/L as $CaCO_3$) and effluent (97 ± 8.5 mg/L as $CaCO_3$) alkalinities at Amherst were not significantly different ($\alpha = 0.05$ and 0.01, StT) during Phase I (Figure 6). Unlike the Mont Vernon system, there was no increase in alkalinity through the unit.

The pH and alkalinity data obtained at Mont Vernon and Amherst during their start up periods is best explained by the influent conditions and the chemical composition of the GAC. It has been shown that coconut shell-based carbons can contain significant quantities of $CaCO_3(s)$ (from the base material) and lime ($Ca(OH)_2$ or CaO) from the activation process (S.J. Shields; Barnebey and Sutcliffe, Corp.; personal communication; 1989). Therefore, the initial rise in pH and alkalinity observed at Mont Vernon can be explained by the dissolution of $CaCO_3(s)$ and/or lime present in the GAC. Further, the theoretical solubility for $CaCO_3(s)$ (Stumm and Morgan, 1981) for the water quality conditions at Mont Vernon and Amherst would predict that rapid dissolution should occur below pH 8 (i.e., at the Mont Vernon and Amherst influent pH's) and that the equilibrium pH of the $CaCO_3(s)$ system following dissolution would be between pH 9.5 and 10.5 which corresponds to the peak effluent pH values observed at both sites.

The differences observed at Mont Vernon and Amherst are likely the result of several factors: (i) influent pH and alkalinity at Amherst were much higher than at Mont Vernon, (ii) the Amherst system contained only 0.85 m^3 of GAC whereas the Mont Vernon system contained 1.33 m^3, and (iii) the average daily flow at Amherst (59 m^3/day) was almost double that of Mont Vernon (31 m^3/day). These factors serve to explain why the pH and alkalinity changes (influent vs. effluent) observed at Amherst where reduced and occurred for a shorter period. Subsequent data for Mont Vernon shows that by Day 50 there were no significant differences between influent and effluent pH or alkalinity. This confirms that the trend observed for Amherst also occurred at Mont Vernon, but due to the greater volume of GAC and lower flowrate, a longer operating period was required to flush the system of the $CaCO_3$ and/or lime.

Temperature and Dissolved Oxygen--

Mont Vernon site--The temperature of the water was not affected by the treatment units, never changing by more than 1.0°C from influent to effluent (Figure 7). The average temperature of the effluent water was 10.9 ± 0.9°C and did not differ significantly ($\alpha = 0.05$ and 0.01, StT) from the influent (11.1 ± 0.7°C). This was expected, in spite of the detention times (0.4 - 1.7 hr) because the pumphouse was not heated. The startup period occurred in October and November when ambient air temperatures in New England are cool.

Dissolved oxygen (DO) in the influent water averaged 10.8 ± 0.73 mg O_2/L during Phase I. A substantial amount of DO was removed on Day 1 (influent = 9.8 mg O_2/L, effluent = 1.5 mg O_2/L) (Figure 7). There was an increase in effluent DO from 1.5 mg O_2/L on Day 1 to 4.5 mg O_2/L on Day 15. However, by

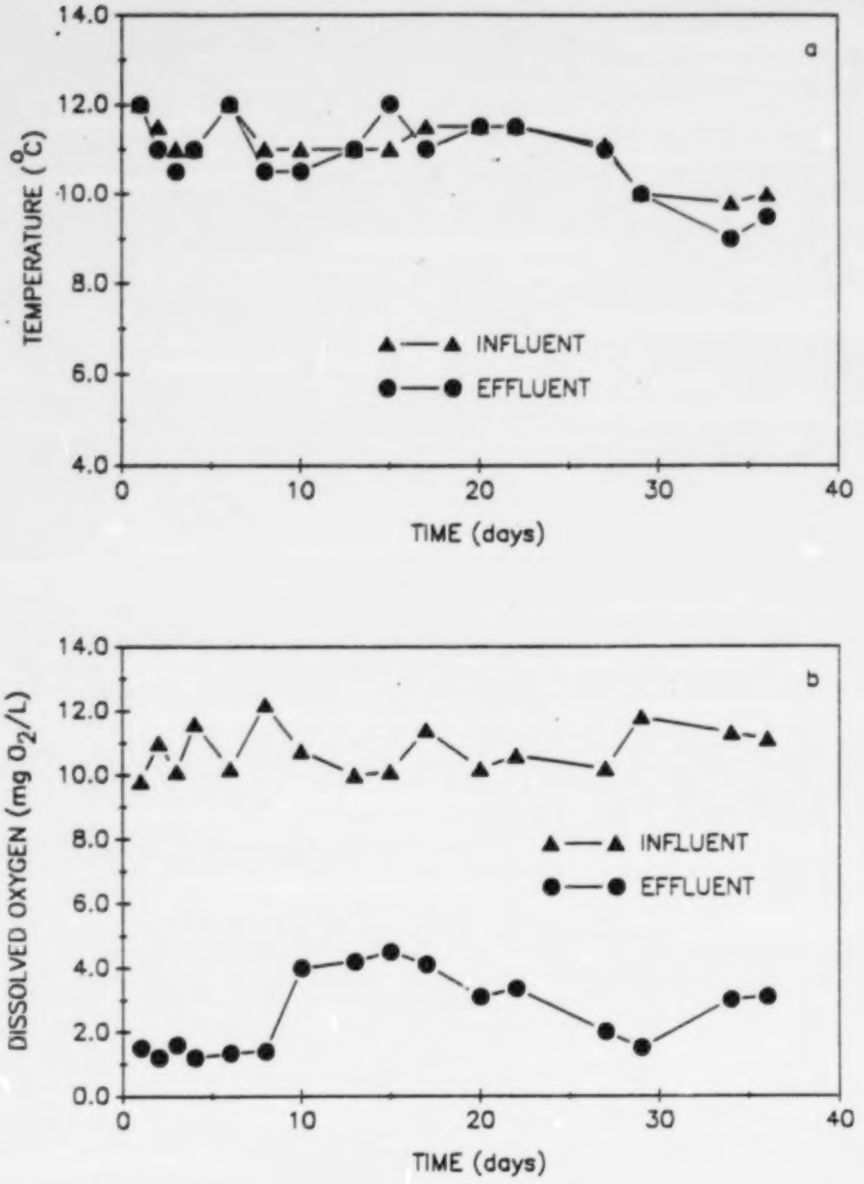


Figure 7. Phase I - Mont Vernon, NH GAC system. Temperature (a) and dissolved oxygen (b) for the first 36 days of operation.

Day 27 the effluent DO concentration decreased to 2.0 mg O_2/L and remained substantially lower than the influent throughout Phase I. Overall, the effluent DO concentration averaged 2.4 ± 1.2 mg O_2/L for the 36 day period of Phase I.

Amherst site--The temperature of the water was not affected by the treatment unit, never changing by more than 1.0°C from influent to effluent (Figure 8). The average temperature of the effluent water (11.7 ± 1.1°C) did not differ significantly ($\alpha = 0.05$ and 0.01, StT) from the influent water temperature of 11.9 ± 1.0°C. The temperature of the water was not expected to change because of the short detention time in the GAC unit ranging from 0.28 to 0.37 hr and the cool ambient air temperatures in the pumphouse during the start-up period (October and November).

Dissolved oxygen in the influent water averaged 4.9 ± 0.67 mg O_2/L during Phase I. During this 32 day period, DO was consistently removed by the units (Figure 8), and the effluent DO concentration averaged 2.8 ± 0.84 mg O_2/L.

Analysis of the temperature and DO data collected during Phase I indicates that both systems were performing as expected. The minimal change in temperature is consistent with results observed for numerous applications of GAC in groundwater treatment. Snoeyink (1983) reported that the effect of temperature on GAC adsorption from groundwater would be minimal compared to other factors.

The DO data can be evaluated by computing an average mass of DO adsorbed per mass of GAC for each system based on average flow, average influent DO and effluent DO concentration and the time of operation. The resulting calculations indicate that 13.9 mg O_2/g GAC were adsorbed during the 36 day period of Phase I at Mont Vernon and 9.1 mg O_2/g GAC were adsorbed during Phase I (32 days) at Amherst. These values compare well to those reported by Prober et al. (1975), who found DO adsorption averaged 4.8 to 21 mg O_2/g GAC for a variety of GAC's tested up to a period of 1,700 hrs (70.8 days). They also reported that GAC exhaustion was not reached during the 70.8 day study. This finding is consistent with results observed at Mont Vernon and Amherst, during Phase I, since neither system demonstrated complete GAC exhaustion with respect to DO (i.e., influent DO = effluent DO).

The differences observed in DO adsorption at Mont Vernon and Amherst are likely the result of the influent DO concentrations and the volume of GAC at each site. Since the driving force (i.e., concentration gradient) of DO adsorption at Mont Vernon was higher than at Amherst, the higher non-equilibrium adsorption of DO observed at Mont Vernon would be expected. Further, Prober et al. (1975) concluded that up to 95% of the DO adsorbed by GAC results from surface reactions with GAC forming carboxylic and other acidic surface groups. Therefore, the fact that Mont Vernon had 1.33 m^3 of GAC (Amherst = 0.85 m^3) may also explain the higher DO adsorption at Mont Vernon.

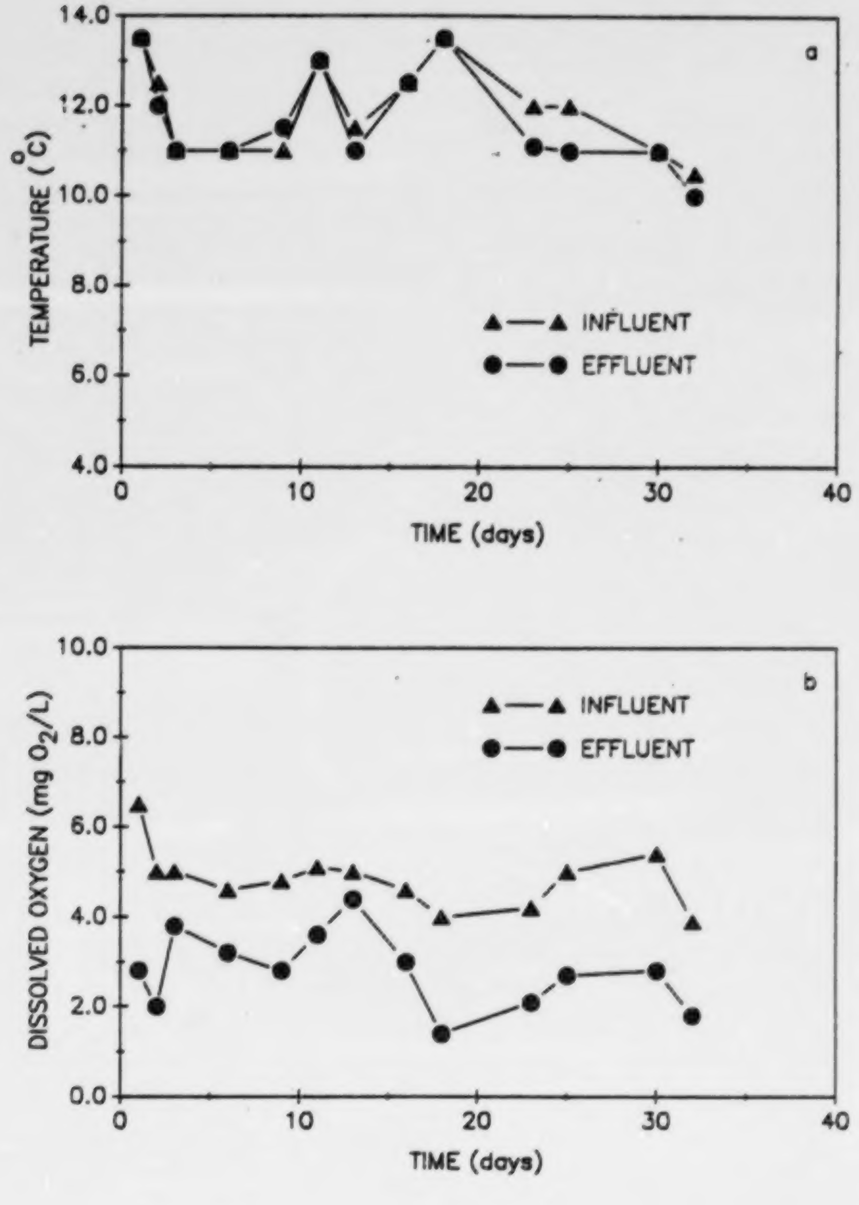


Figure 8. Phase I - Amherst, NH GAC system. Temperature (a) and dissolved oxygen (b) for first 32 days of operation.

Turbidity--

Mont Vernon site--During Phase I, the average influent turbidity was 0.17 ± 0.05 NTU (Figure 9). The effluent turbidity during this period (0.12 ± 0.04 NTU) was significantly less than the influent (α = 0.05 and 0.01, StT). In a survey of water treatment plants in the U.S. that used GAC, influent turbidities of 1.1 to 7.0 NTU were decreased to 0.1 to 0.5 NTU through the units (Graese, et al. 1987). Although a slight reduction in turbidity by the Mont Vernon system was observed, the low influent turbidity, typical of many groundwaters, made hydraulic headloss through the GAC bed negligible during Phase I.

Amherst site--The average Phase I influent turbidity was 1.75 ± 0.98 NTU (Figure 10). This is significantly higher (α = 0.05 and 0.01, StT) than Mont Vernon's. Some of this difference may have been a function of the iron precipitates observed entering the Amherst GAC system from the atmospheric storage tank. The effluent turbidity during this period was 0.35 ± 0.11 NTU, which was significantly less than the influent turbidity (α = 0.05 and 0.01, StT). Graese et al. (1987) surveyed a GAC water treatment plant in the United Kingdom (GAC type and volume unknown) whose influent turbidity of 1.5 NTU decreased to 0.32 NTU through the unit, similar to the trend observed in Amherst. Despite fluctuations in influent turbidity, the GAC unit produced a relatively constant effluent. The effluent turbidity at Amherst may have been higher than Mont Vernon's because the GAC volume was lower and raw water turbidity and flow were higher. Despite the higher turbidity reduction at Amherst, headloss was not considered significant during Phase I.

Flowrate--

Variations in water flowrate are significant in radon removal, because removal is controlled by the empty bed contact time (EBCT) which is a function of contactor volume and flowrate (Lowry and Brandow, 1985). The average flowrate was determined from the total volume of water passed through the GAC systems over periods between sampling. During Phase I, measurements were taken daily or every few days, which was considered adequate for this study. However, in small systems where rapid changes in flow can significantly affect the data, flows should be taken immediately before and after each sampling event for a more accurate determination of mass radon removals.

Mont Vernon site--The average flow during Phase I at Mont Vernon was 24 ± 4 m^3/day (Figure 11) yielding an average EBCT of 78.5 min. The GAC system was designed for an average flow of 25 m^3/day (EBCT=77.9 min).

Amherst site--The average flow at the Amherst facility during Phase I was 59 ± 3.2 m^3/day (Figure 12) which corresponds to an average EBCT of 20.6 min. Flow data was not collected prior to Day 6 because there was a delay in installation of the monitoring equipment. This flowrate was much higher than that monitored at Mont Vernon and higher than the anticipated design flow of 34 m^3/day (EBCT=35.5 min). The flowrate was fairly constant over the period, except when the GAC unit was shutdown by the operator (Days 4 to 8).

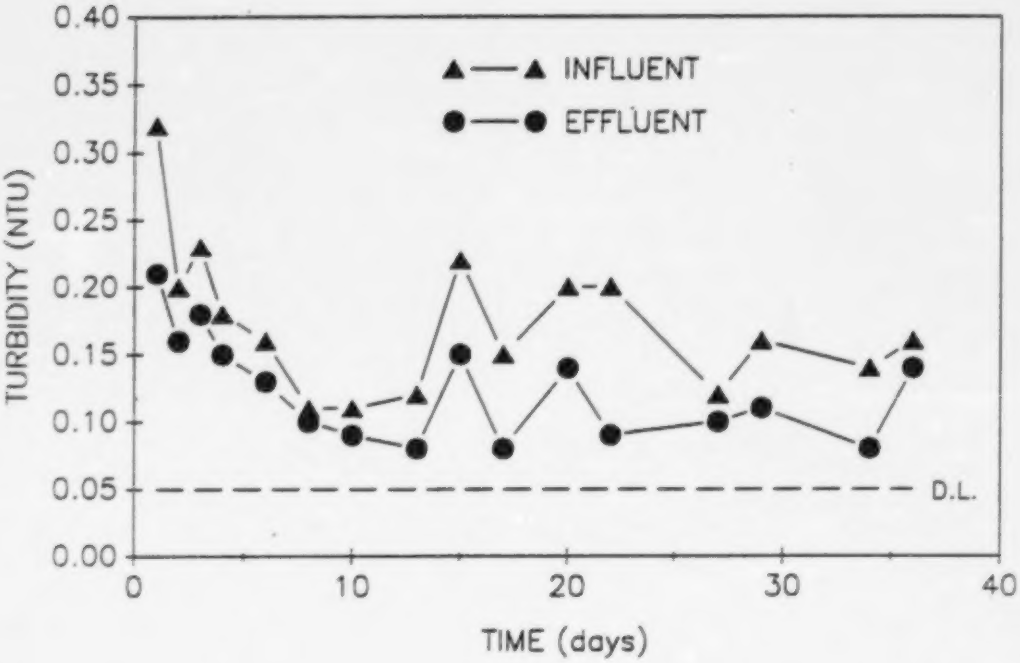


Figure 9. Phase I - Mont Vernon, NH GAC system. Turbidity during first 36 days of operation (detection limit = 0.05 NTU).

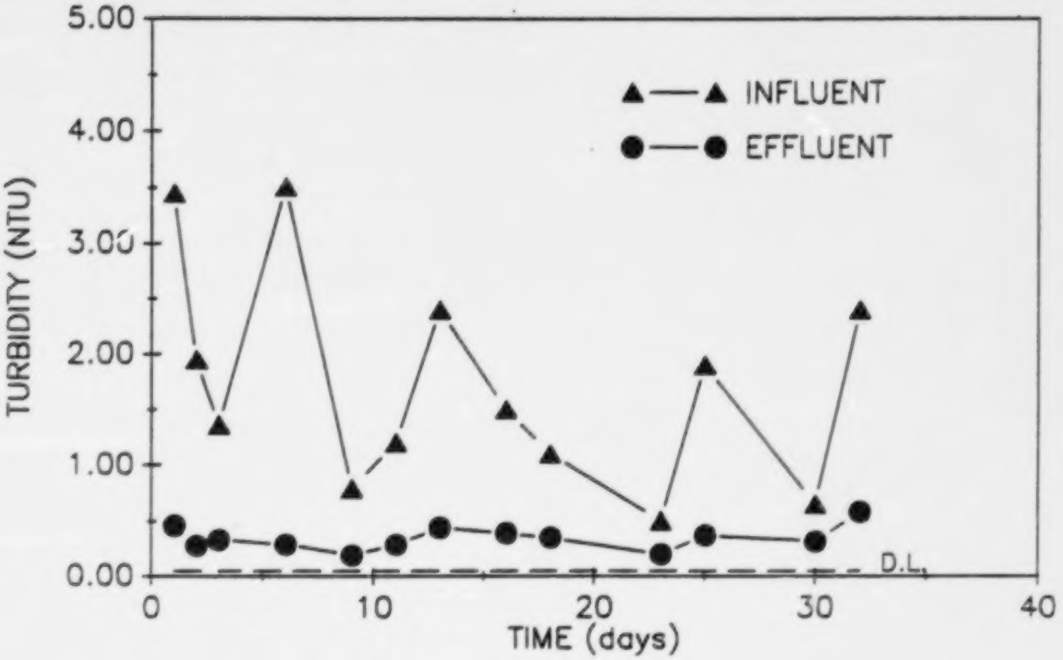


Figure 10. Phase I - Amherst, NH GAC system. Turbidity during first 32 days of operation (detection limit = 0.05 NTU).

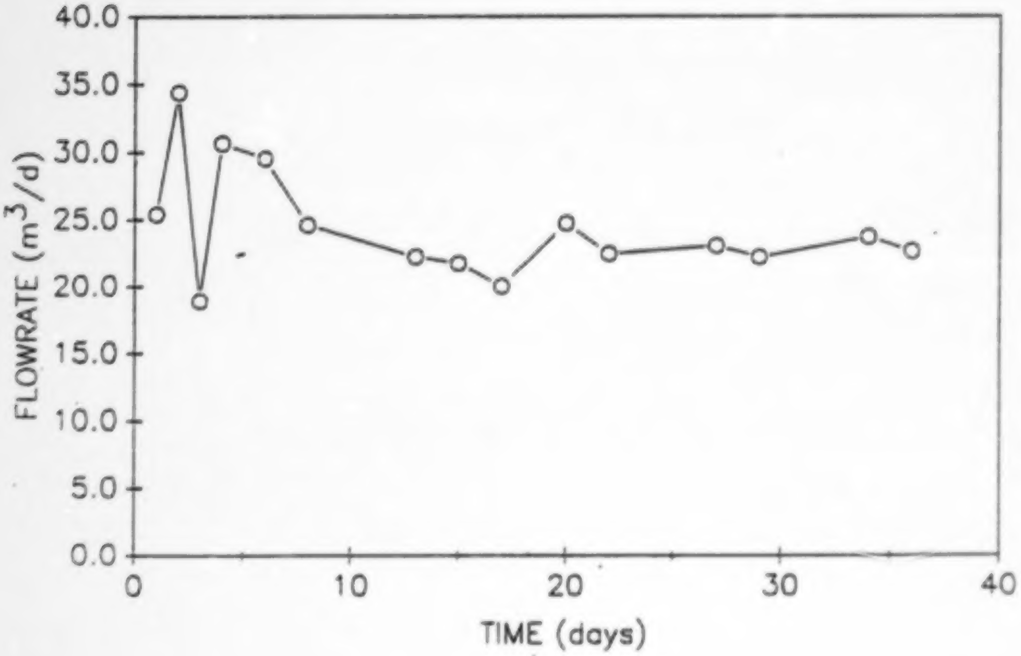


Figure 11. Phase I - Mont Vernon, NH GAC system. Water flowrate through GAC during first 36 days of operation.

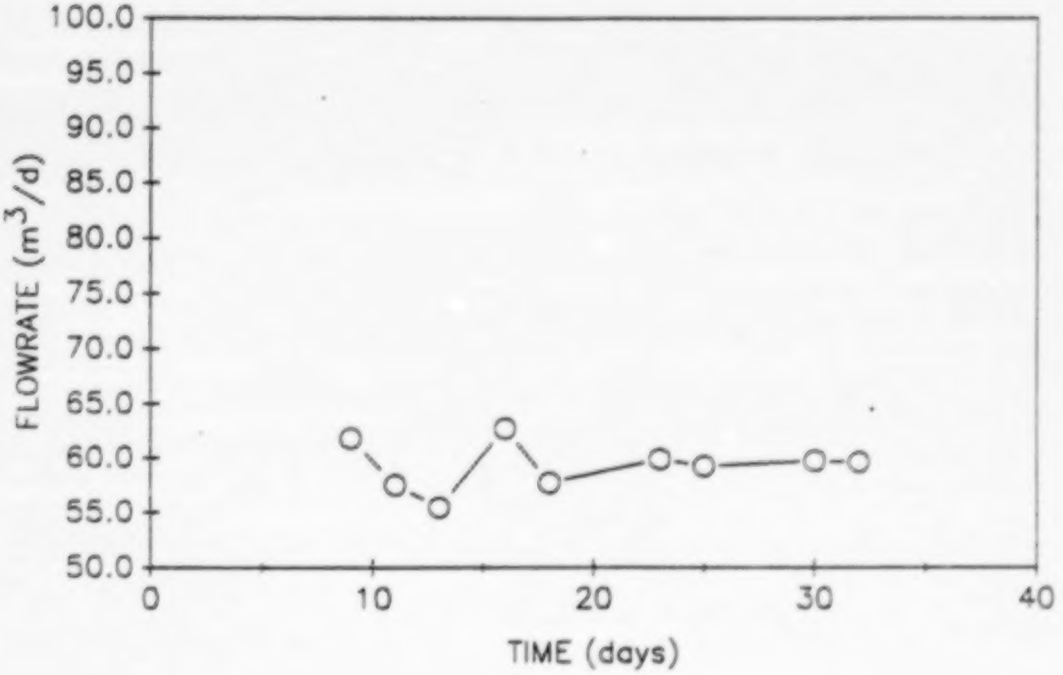


Figure 12. Phase I - Amherst, NH GAC system. Water flowrate from Day 6 to Day 32 (metering equipment not installed until Day 5).

Radon--

Mont Vernon site--The GAC system at Mont Vernon was designed to treat an influent radon activity of 155,000 pCi/L and to achieve a 98% radon removal efficiency once steady-state operation was achieved (predicted effluent activity = 2,860 pCi/L). The average influent radon activity during Phase I was 249,900 ± 45,800 pCi/L.

Figure 13 presents radon data collected from each port during Phase I. All of the radon applied was removed in the first unit (GAC #1) during the first few days of operation. The radon removal wave front moved through the units over time, eventually breaking through into the effluent by Day 25. Prior to that the radon activity was below the detection limit (138 pCi/L) in the effluent from GAC #2. By Day 36, the effluent activity from GAC #2 was 2530 pCi/L.

Based on the adsorption data for GAC #2, this unit was not at steady state with respect to radon removal by the end of Phase I. The data from GAC #1 indicated that it was near steady state conditions within approximately 20 days, considering the varying water flowrate and influent radon activity inherent in a small community water supply. Using a radon adsorption model developed for GAC by Lowry (1985), the influent and effluent radon activity and the averaged flowrate over the period, the steady state radon removal rate constant (K_{ss}) was determined for comparative purposes. The K_{ss} for GAC #1 averaged 3.3 hr^{-1} (ranging from 2.7 hr^{-1} in the top 0.14 m^3 of GAC to 4.9 hr^{-1} in the bottom 0.15 m^3) once secular equilibrium was achieved between radon and its progeny. The design K_{ss} used for this carbon at water temperatures in the range 6-10°C was 3.02 hr^{-1} (Lowry and Lowry, 1987), which was similar to the average value observed. It is well documented for conventional pollutants (Weber, 1972; Snoeyink, 1983; Crittenden, 1987) that GAC adsorption is affected by column hydraulics and site specific raw water quality conditions. Therefore, variations in K_{ss} would be expected as column hydraulics and raw water quality change. As predicted by Lowry (1985), the GAC unit dampened changes in loading once it was at steady state.

Amherst site--The GAC system at Amherst was designed to treat an average influent of 39,750 pCi/L and provide a steady-state effluent of 6,280 pCi/L (84% radon removal) at a flowrate of 34 m^3/day. The average influent radon activity during Phase I was 56,600 ± 11,100 pCi/L (Figure 14).

As observed at Mont Vernon in GAC #1 (0.57 m^3), the GAC system at Amherst (0.85 m^3) came to a steady state condition fairly rapidly (~15 days). This is similar to observations ranging from 10-22 days for other GAC units (Lowry et al., 1987; Lowry and Brandow, 1985; Reed, 1989). The radon appeared in the effluent within a few days of startup and increased to a steady state average of 7,971 ± 738 pCi/L after 15 days of operation. Fluctuations in influent loading were also dampened in the Amherst system, however, to a lesser extent than at Mont Vernon where there was a greater volume of GAC.

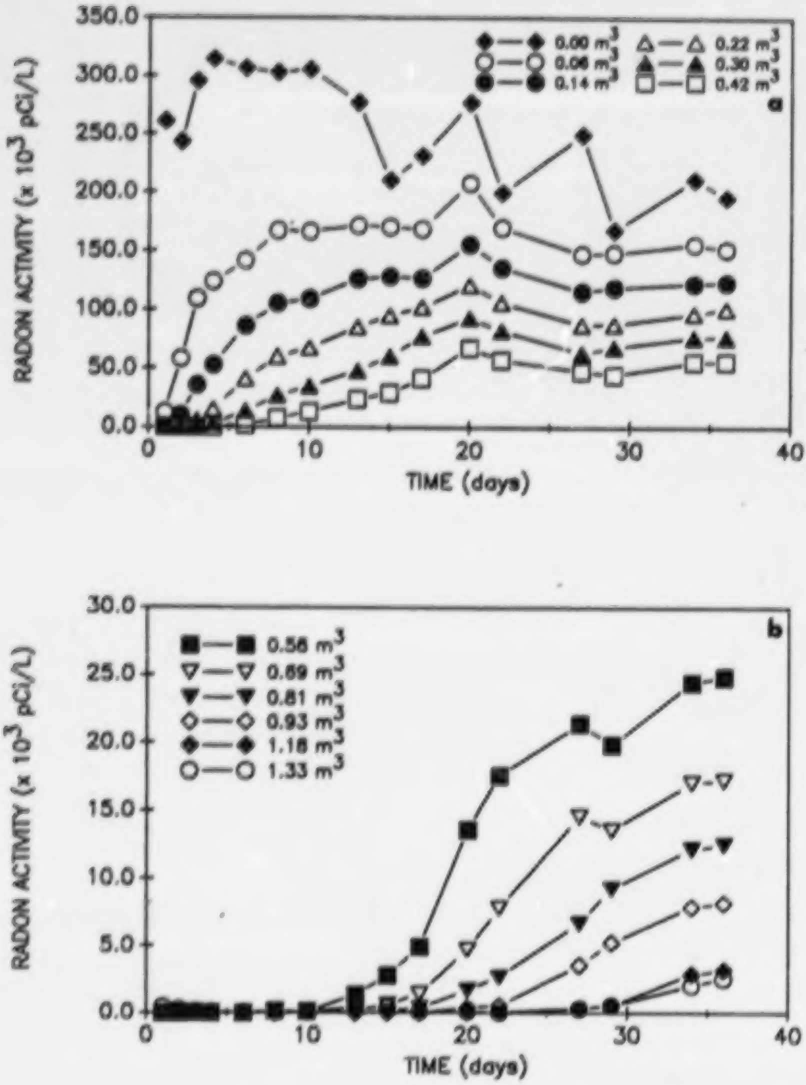


Figure 13. Phase I - Mont Vernon, NH GAC system. Radon activity during first 36 days of operation for GAC #1 (a) and GAC #2 (b). Note difference in scales on ordinate in (a) and (b).

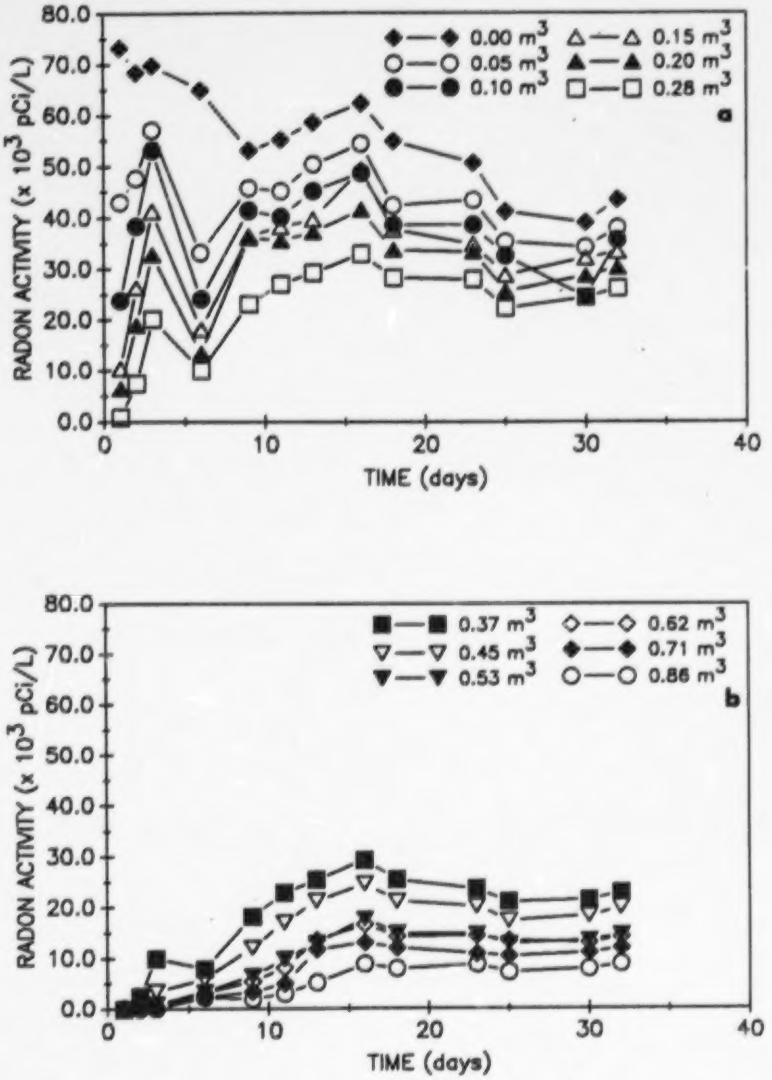


Figure 14. Phase I - Amherst, NH GAC system. Radon activity during first 32 days of operation for top 0.28 m³ (a) and remaining 0.58 m³ of GAC (b).

For the Amherst GAC system, the K_{ss} averaged 5.8 ± 1.44 hr^{-1} in the bottom 0.41 m^3 (when the radon loading and flowrate were fairly constant) once the unit was at secular equilibrium. These K_{ss} values are substantially higher than those predicted (3.02 hr^{-1} at 6-10°C) for this GAC (Lowry and Lowry, 1987) and may be a function of the column hydraulics and/or the raw water quality at the Amherst site.

Gamma/Beta Emissions--

Mont Vernon site--Results of the Geiger Müeller (GM) survey meter readings taken at the surface of the GAC units during Phase I are presented in Figure 15. The GM survey meter measures both gamma and beta emissions when the detector is unshielded. Two of the short-lived progeny of radon, lead-214 and bismuth-214, emit fairly high energy gamma radiation. This gamma activity penetrates the fiberglass tanks and is detected by the survey meter. For GAC #1, these emissions were fairly constant within 20 to 30 days of start-up corresponding to the radon activities observed. Gamma/beta emissions from GAC #2 increased after Day 5 corresponding to the breakthrough of radon from GAC #1. Figure 16 shows the correlation between the radon activity at the closest port to the gamma/beta emissions measurements during Phase I for GAC #1.

Amherst site--The GM survey meter reading results for Amherst are presented in Figure 17. The data from the top of the unit show some fluctuation, which was dampened through the system. Again, the radon activity was correlated with the gamma/beta emissions (Figure 18), however at this site a non-linear relationship appears to provide the best fit of the data.

Microbial Numbers--

These analyses were not performed during Phase I.

Iron and Manganese--

Mont Vernon site--On several occasions during the beginning of operation, the effluent concentrations of both iron and manganese exceeded the influent concentration (Figure 19). Thereafter, the influent iron and manganese were usually higher than the effluent from the GAC system. After Day 17, the majority of the influent and effluent data were at or near the detection limit (iron = 0.06 mg/L, manganese = 0.02 mg/L), so changes in iron and manganese could not be determined through the GAC.

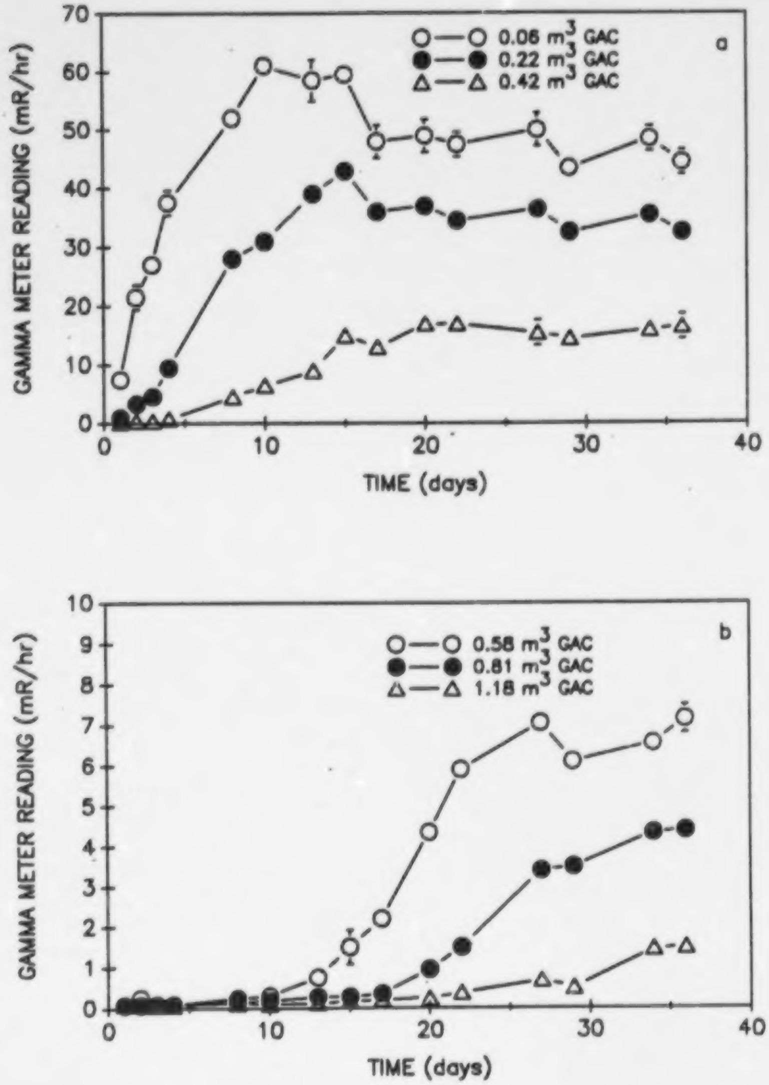


Figure 15. Phase I - Mont Vernon, NH GAC system. Gamma/beta emission measurements taken on contactor surfaces during first 36 days of operation for GAC #1 (a) and GAC #2 (b). Note difference in scale.

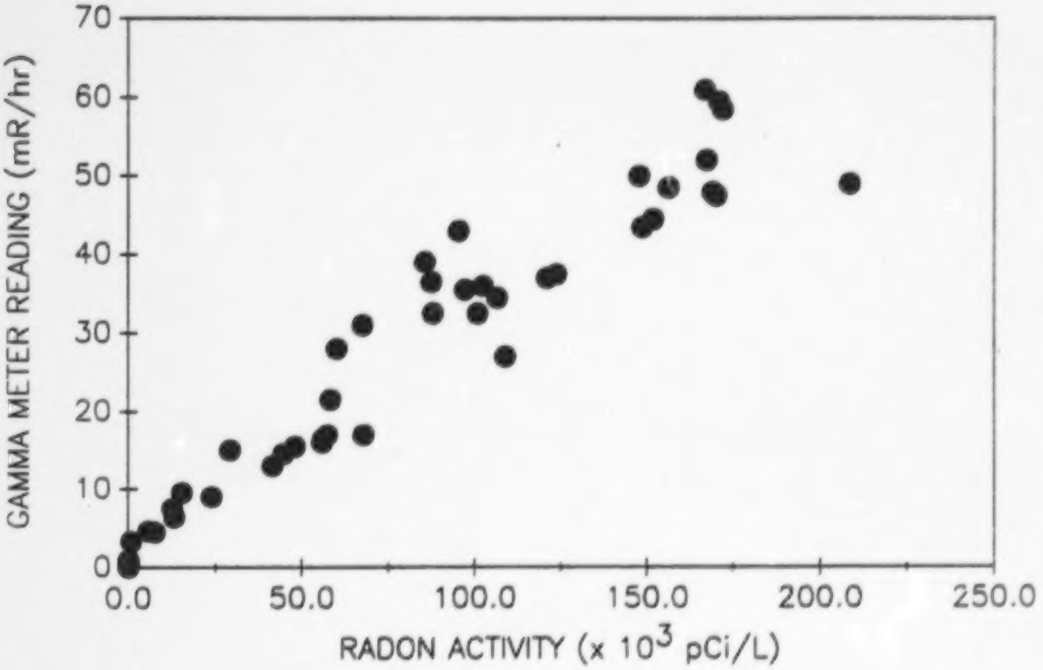


Figure 16. Phase I - Mont Vernon, NH GAC system. Gamma/beta emission measurements taken on the surface of GAC #1 as a function of influent radon activity to sections of GAC #1.

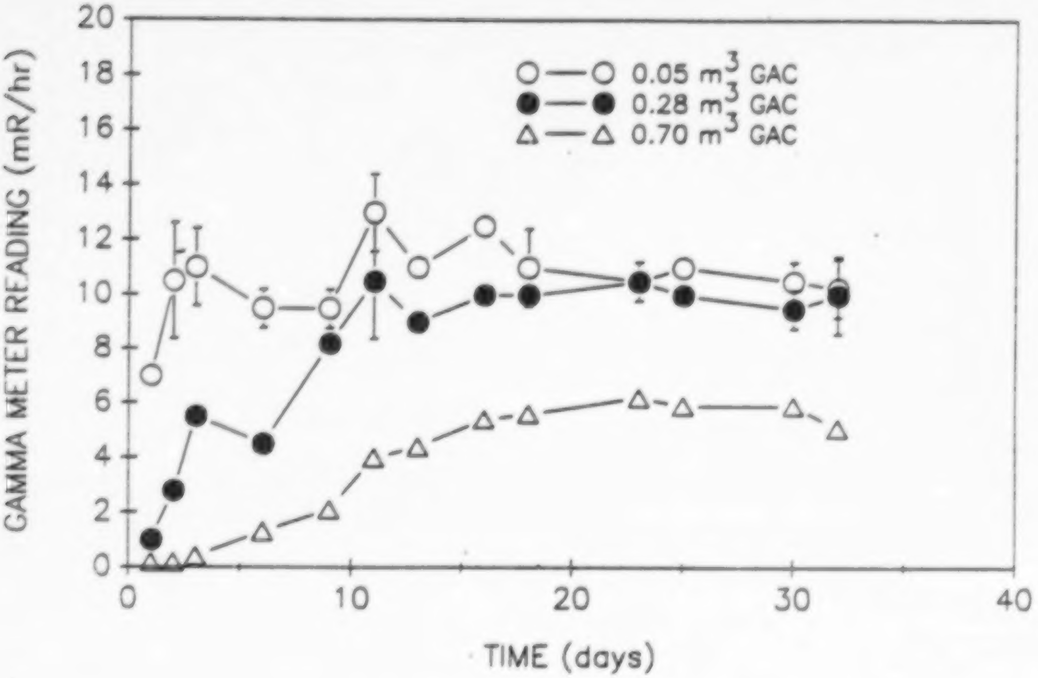


Figure 17. Phase I - Amherst, NH GAC system. Gamma/beta emission measurements taken on the contactor surface during first 32 days of operation.

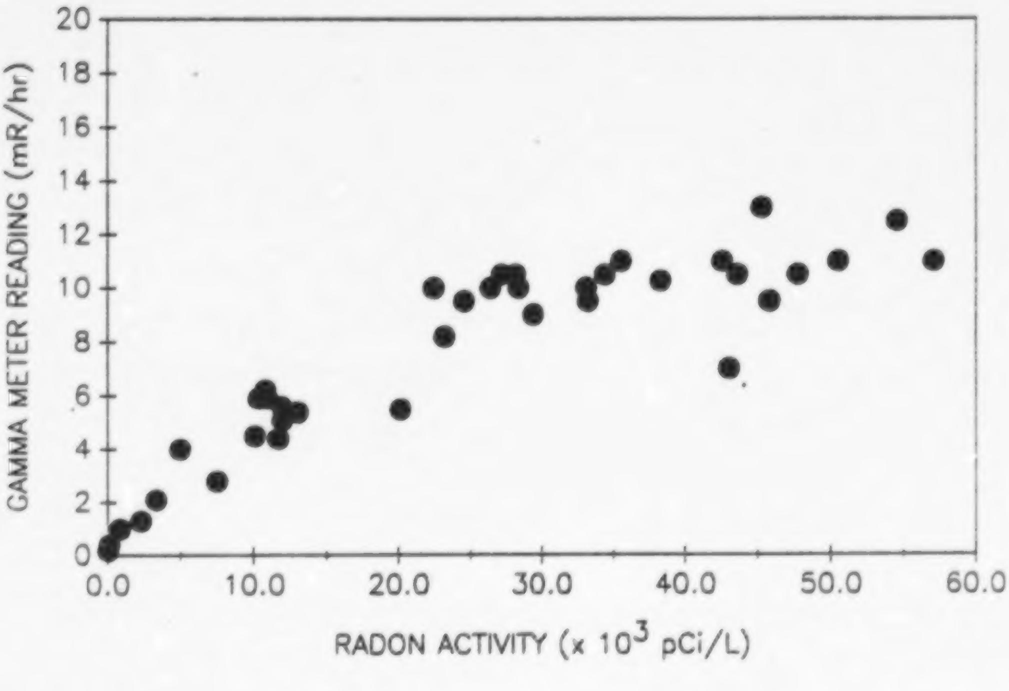


Figure 18. Phase I - Amherst, NH GAC system. Gamma/beta emission measurements taken on the contactor surface as a function of influent radon activity to sections of the GAC system.

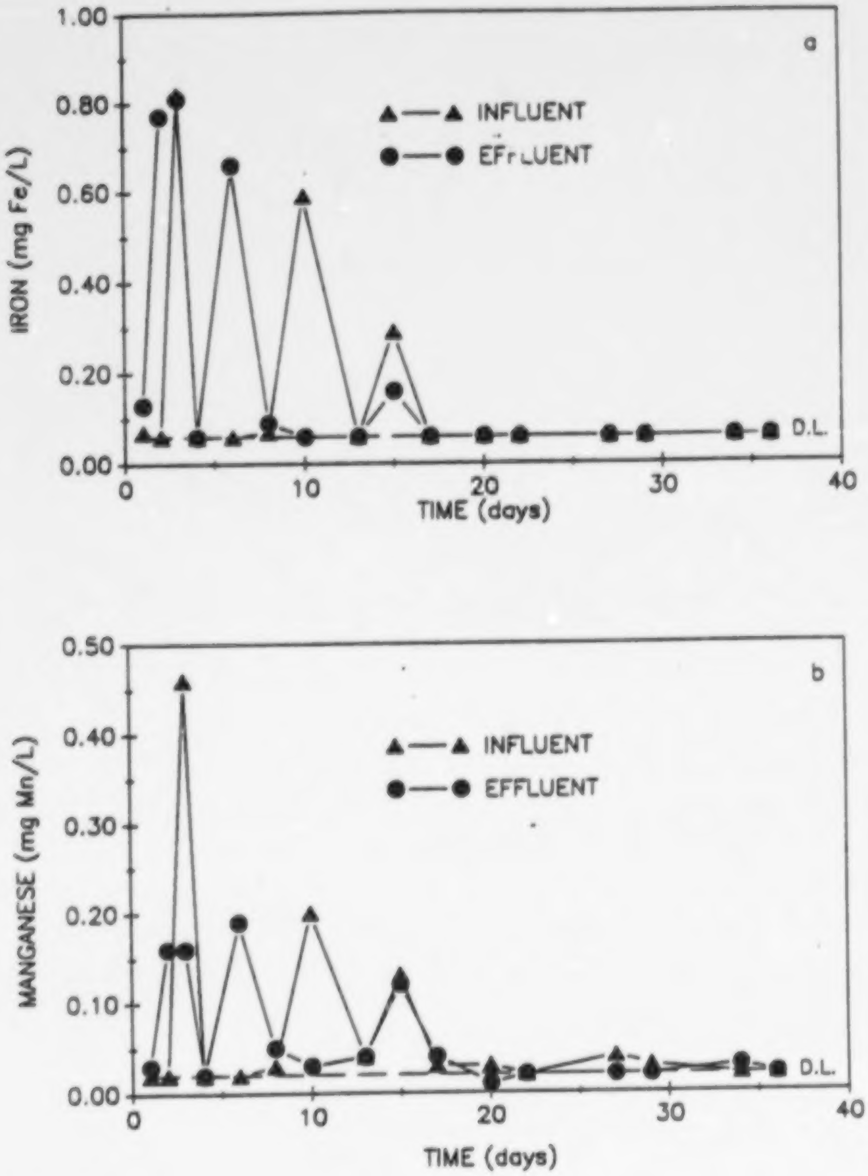


Figure 19. Phase I - Mont Vernon, NH GAC system. Iron (a) and manganese (b) data for the first 36 days of operation. (Detection limits: iron = 0.06 mg/L and manganese = 0.02 mg/L).

Amherst site--Influent iron concentrations were significantly higher (α = 0.05 and 0.01, StT) in the Amherst water than the Mont Vernon water (Figure 20). There was no significant difference (α = 0.05 and 0.10, StT) in raw water manganese concentrations between the sites (Figure 20). The average influent iron and manganese concentrations at Amherst were 0.50 ± 0.30 mg Fe/L and 0.09 ± 0.08 mg Mn/L, respectively. The average effluent concentrations were 0.19 ± 0.29 mg Fe/L and 0.06 ± 0.07 mg Mn/L. There was a significant difference at α = 0.05 and 0.02 (StT) in influent and effluent iron, but not at α = 0.01. Removal was observed for both metals through the GAC system which has been observed in other GAC units (Graese, 1987).

Phase II - Long-Term Monitoring

During Phase II, the GAC system at Mont Vernon was monitored weekly for 16 weeks, bi-weekly for 24 weeks and then monthly for two months. Between September 17, 1987 (Day 344) and October 15, 1987 (Day 372), a new well with a higher yield was drilled and replaced the existing wells as the water supply. The water in the new well contained less radon and DO, but considerably more uranium and radium and had a higher pH and alkalinity than the original water supply (Table 5). These data support observations by Boudette (NH State Geologist; Durham, NH; personal communication, 1988) that there may be considerable variation in radon activity in groundwater from neighboring wells, largely as a function of their depth within the aquifer.

The GAC system at Amherst was monitored weekly for 5 weeks and then bi-weekly for 12 weeks. Several problems were encountered with the water supply system after Day 122. The major problem was the very low well yield. In addition, the individual employed to monitor the water supply system repeatedly bypassed the GAC unit by mistake. As a result, the system was abandoned after Day 122.

pH and Alkalinity--

Mont Vernon site--Alkalinity and pH did not differ significantly (α = 0.05 and 0.01, StT) as the water flowed through the GAC system (Figure 21). The alkalinity of the influent water decreased from 49.5 mg/L as $CaCO_3$ on Day 36 to 26.3 mg/L as $CaCO_3$ on Day 43. A similar decrease was observed in the effluent alkalinity indicating that the GAC system was not affecting this parameter. Figure 22 shows typical profiles of pH and alkalinity through the GAC units on several days. Little difference in either parameter was observed from port to port. Day 372 reflects the change in pH and alkalinity resulting from the new water supply.

The continuous monitoring data for pH and alkalinity at Mont Vernon supports the hypothesis that the initial increase in those parameters during Phase I was related to the alkaline bearing residues in the GAC. As predicted by that hypothesis, and observed with the Amherst data, the alkaline material was eventually leached out (solubilized) and the influent and effluent pH and alkalinity were no longer affected by the GAC.

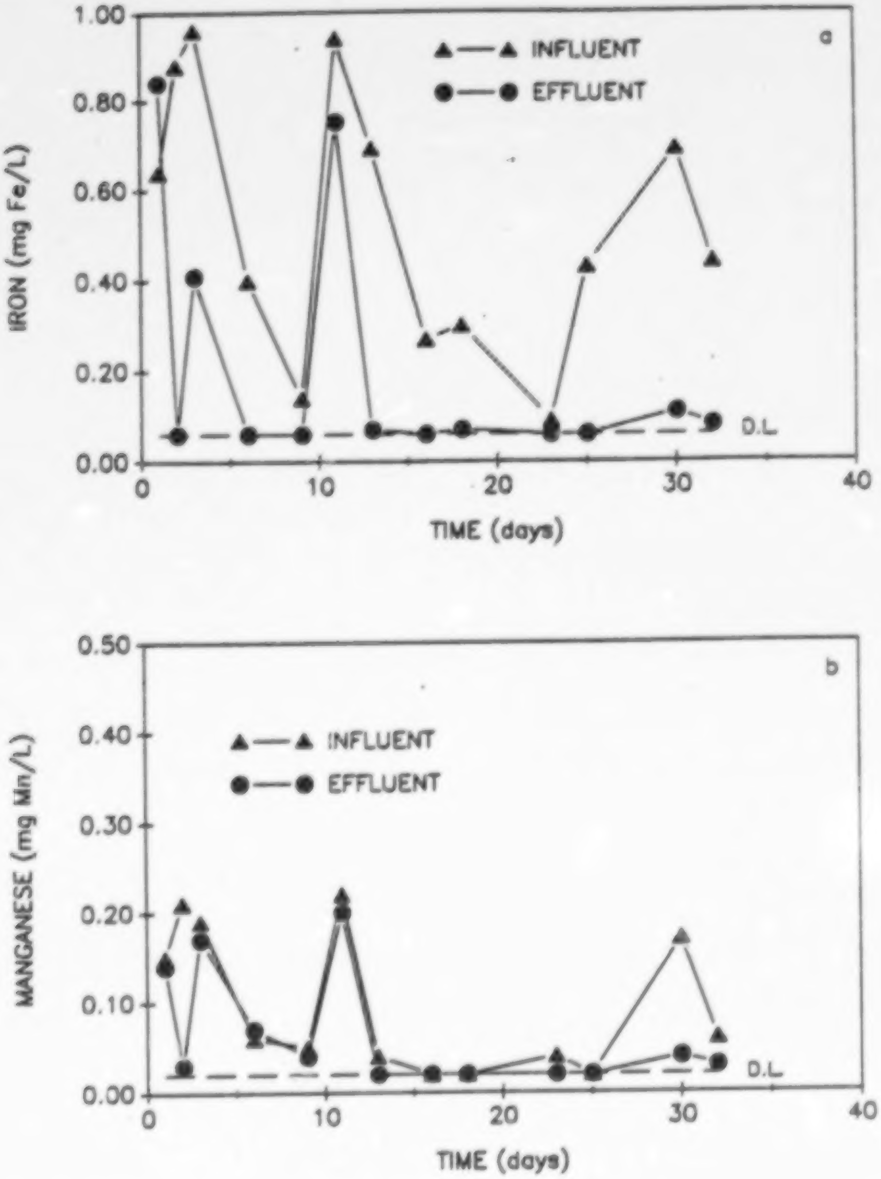


Figure 20. Phase I - Amherst, NH GAC system. Iron (a) and manganese (b) for the first 32 days of operation. (Detection limits iron = 0.06 . mg/L and manganese = 0.02 mg/L).

TABLE 5. MONT VERNON WATER SUPPLY ORIGINAL WELLS VS. NEW WELL

	Original Wells*	New Well⁺
Radon (pCi/L)	210,500 ± 41,400	68,900 ± 1,400
pH	6.5 ± 0.2	7.7 ± 0.03
Temperature (°C)	10.2 ± 1.8	10.7 ± 1.5
Alkalinity (mg/L as $CaCO_3$)	22 ± 1.8	72 ± 3.7
Dissolved Oxygen (mg O_2/L)	10.3 ± 0.5	2.7 ± 0.3
Turbidity (NTU)	0.09 ± 0.05	0.14 ± 0.09
Microbial Populations (CFU/100 ml)	70,200 ± 99,000	52,300 ± 13,400
Iron (mg/L)	BDL**	BDL
Manganese (mg/L)	BDL	BDL
Uranium (pCi/L)	24.6 ± 3.6	528.7 ± 9.0
Radium (pCi/L)	1.4 ± 0.1	> 19⁺⁺

* Based on 27 sampling events.

⁺ Based on 3 sampling events.

** BDL = Below detection limit (0.06 mg Fe/L; 0.02 mg Mn/L).

⁺⁺ Initial sample dilution insufficient.

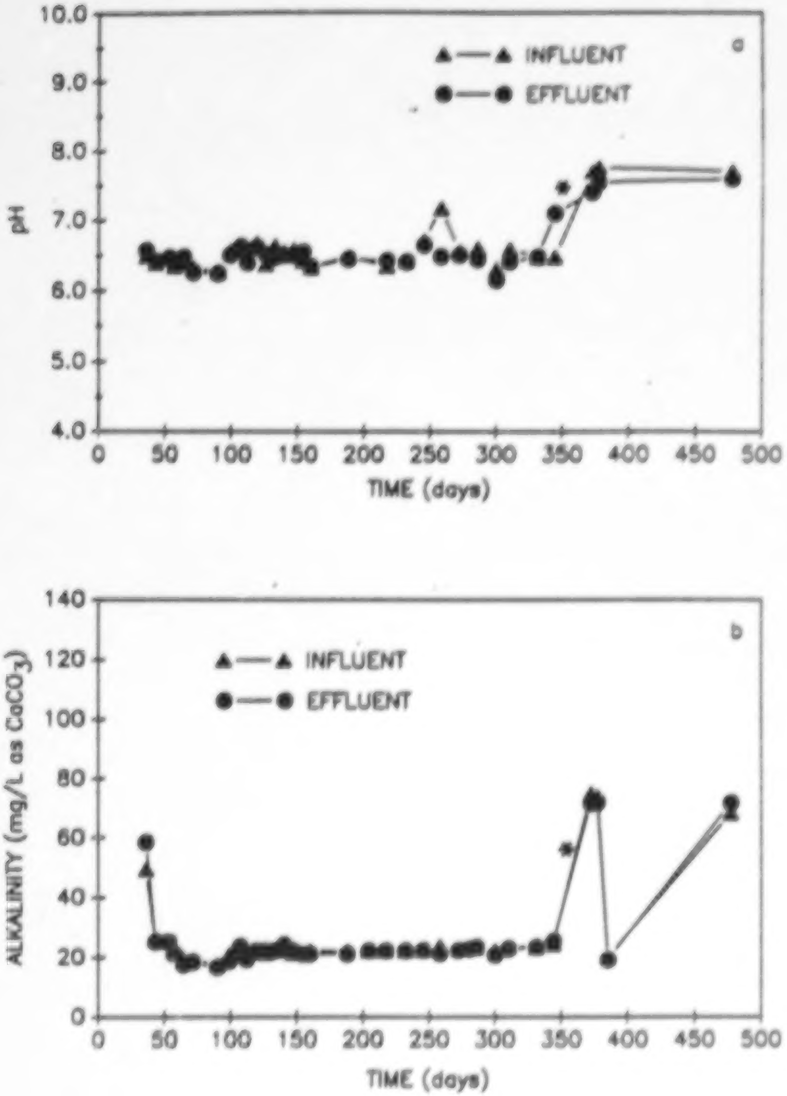


Figure 21. Phase II - Mont Vernon, NH GAC system. pH (a) and alkalinity (b) through 477 days of operation. (* new well)

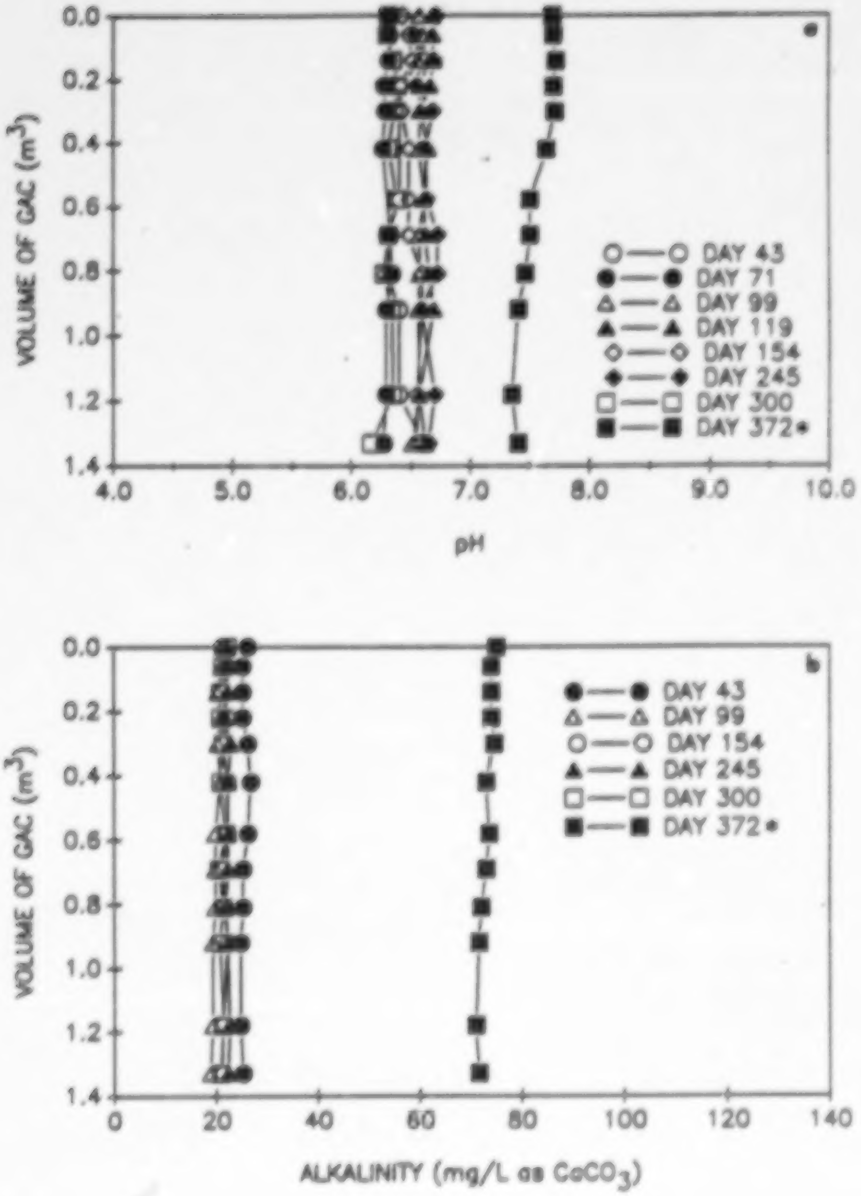


Figure 22. Phase II - Mont Vernon, NH GAC system. Representative profiles of pH (a) and alkalinity (b) through the GAC system. (*new well)

Amherst site--As with the Mont Vernon system, pH and alkalinity were not significantly (α = 0.05 and 0.01, StT) affected as the water passed through the system (Figure 23). Little or no change was observed as the water passed through the unit on most days (Figure 24). The alkalinity of the influent and effluent water decreased at the start of Phase II from 104 ± 10 mg/L as $CaCO_3$ on Day 32 to 55 ± 10 mg/L as $CaCO_3$ on Day 39. This was probably the result of changes in groundwater quality. On Day 39, there was some fluctuation in the lower portion of the unit raising the alkalinity 4 mg/L as $CaCO_3$ (Figure 24), however, there was generally no change in alkalinity within the unit. As previously discussed, additional changes in these parameters were not expected after Phase I, except as a function of variation in influent water quality.

Temperature and Dissolved Oxygen--

Mont Vernon site--Temperature did not differ significantly (α = 0.05 and 0.01, StT) as the water flowed through the GAC units at Mont Vernon (Figure 25). The changes in temperature followed a seasonal pattern, increasing in the spring and summer months and decreasing in the fall and winter. However, temperatures remained in a relatively small range (7.0-13.5°C), typical of northern New England groundwater (Johnson, 1980).

DO began to approach influent concentrations near Day 100 (Figures 25 and 26). However, a fraction of the DO (12.8 to 18.5%) was consistently removed by the GAC units throughout the study.

Amherst site--The temperature of the water was not significantly (α = 0.05 and 0.10, StT) affected as it passed through the system (Figure 27). Fluctuations (4.0 to 13.5°C) were typical for northern New England groundwater and probably represented changes in groundwater temperature and the length of atmospheric storage prior to sampling.

DO continued to be removed in a pattern similar to Phase I (Figure 27). The influent DO in the Amherst water supply was more variable than that at Mont Vernon. The influent and effluent DO were significantly different (α = 0.05 and 0.01, StT) until Day 86, thereafter they were not (α = 0.05 and 0.10, StT). After Day 108, the GAC appeared to approach saturation indicating exhaustion of the adsorptive capacity was occurring (Figure 28).

The continuous monitoring data for DO concentration at both Mont Vernon and Amherst is consistent with the work of Prober et al. (1975), who found that the amount of DO adsorbed per gram of GAC decreased with time, but complete breakthrough was not observed after 70.8 days of operation.

It is hypothesized that the continued DO removal by the GAC may be the result of chemical reactions with the GAC surface, chemical reactions with reduced species adsorbed onto the GAC, and/or microbial activity (i.e., aerobic respiration) within the GAC unit. The fact that the Amherst GAC unit experienced DO saturation whereas the Mont Vernon unit did not, supports the theory that effluent DO is being controlled by reactions with the GAC surface

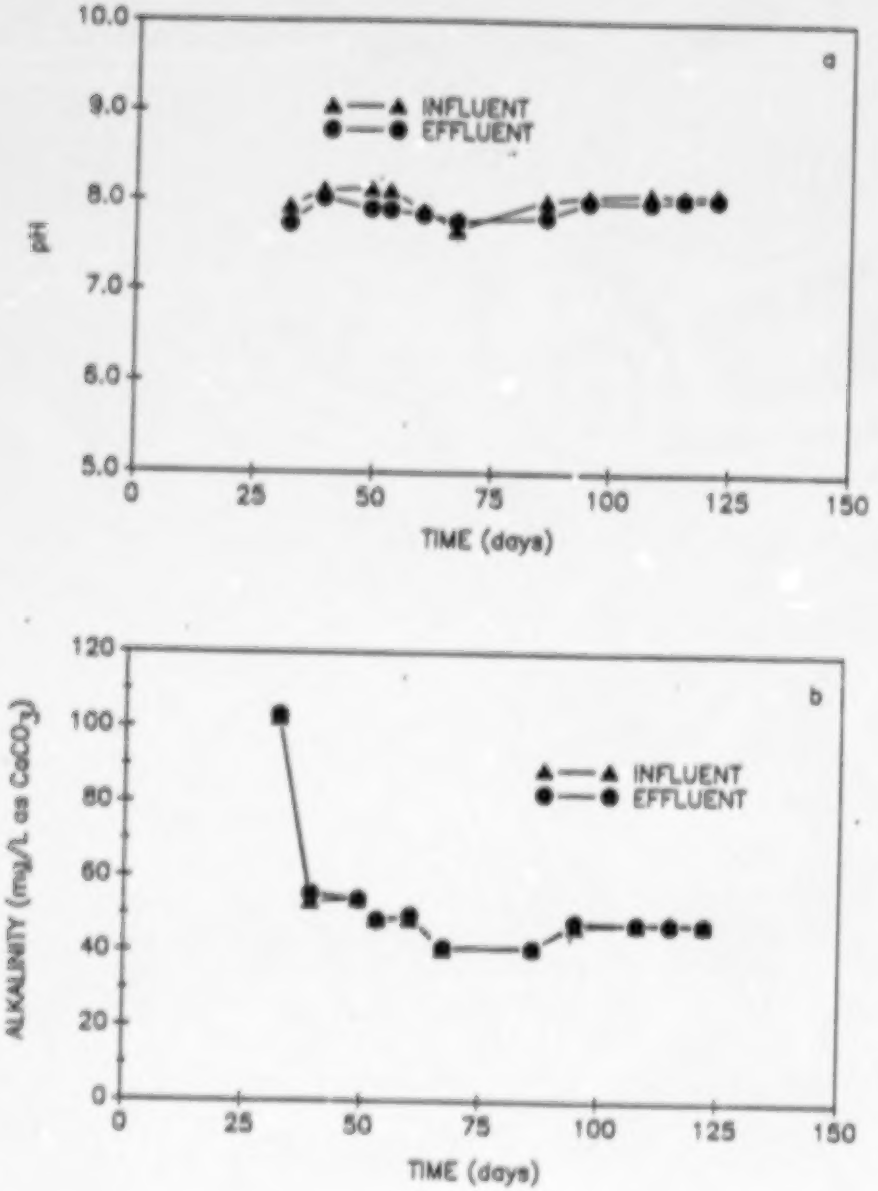


Figure 23. Phase II - Amherst, NH GAC system. pH (a) and alkalinity (b) through 122 days of operation.

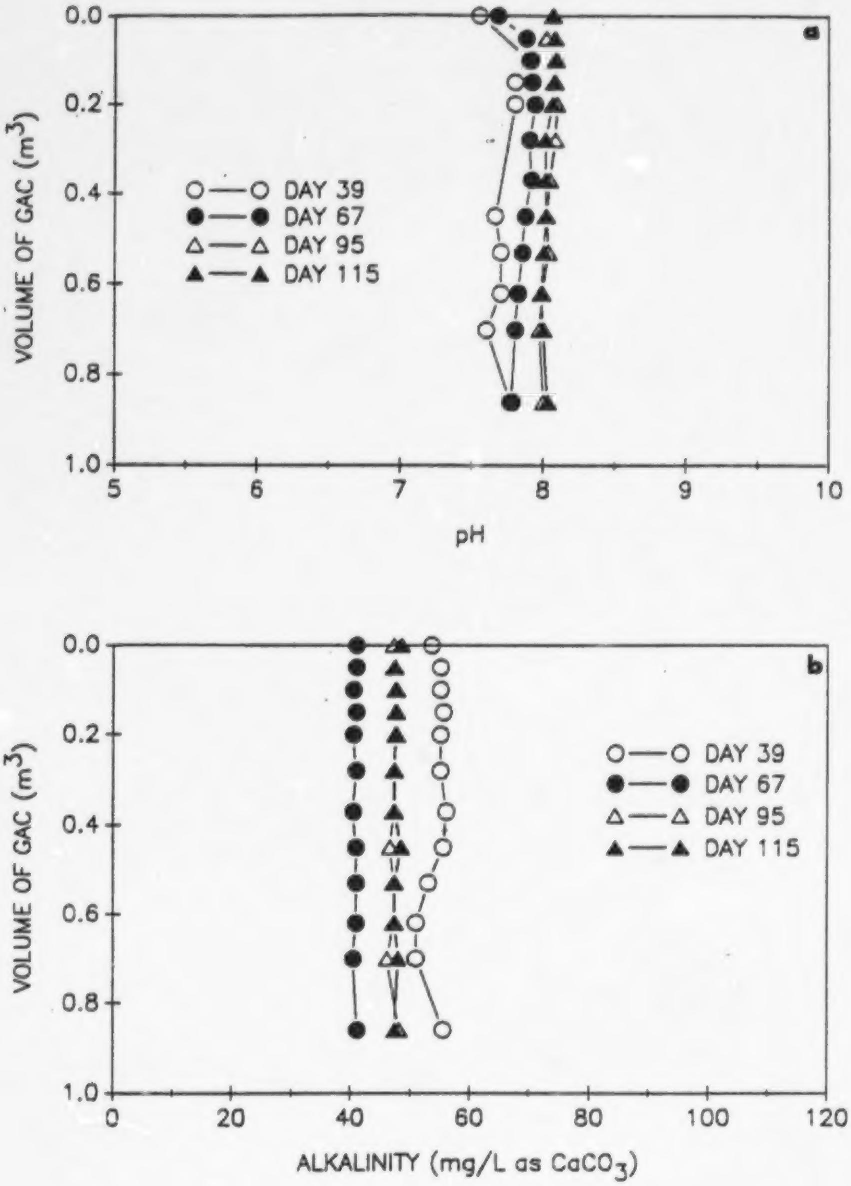


Figure 24. Phase II - Amherst, NH GAC system. Representative profiles of pH (a) and alkalinity (b) through the GAC system.

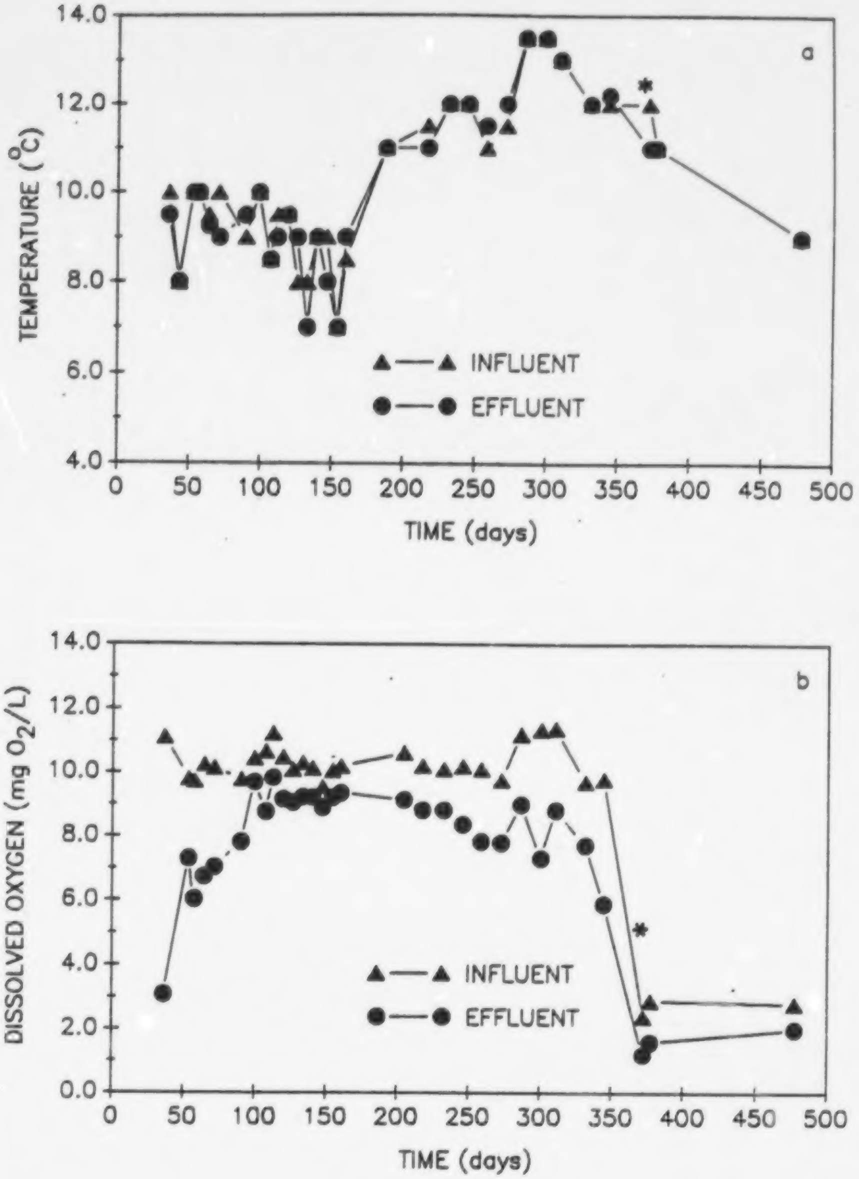


Figure 25. Phase II - Mont Vernon, NH GAC system. Temperature (a) and dissolved oxygen (b) through 477 days of operation. (* new well)

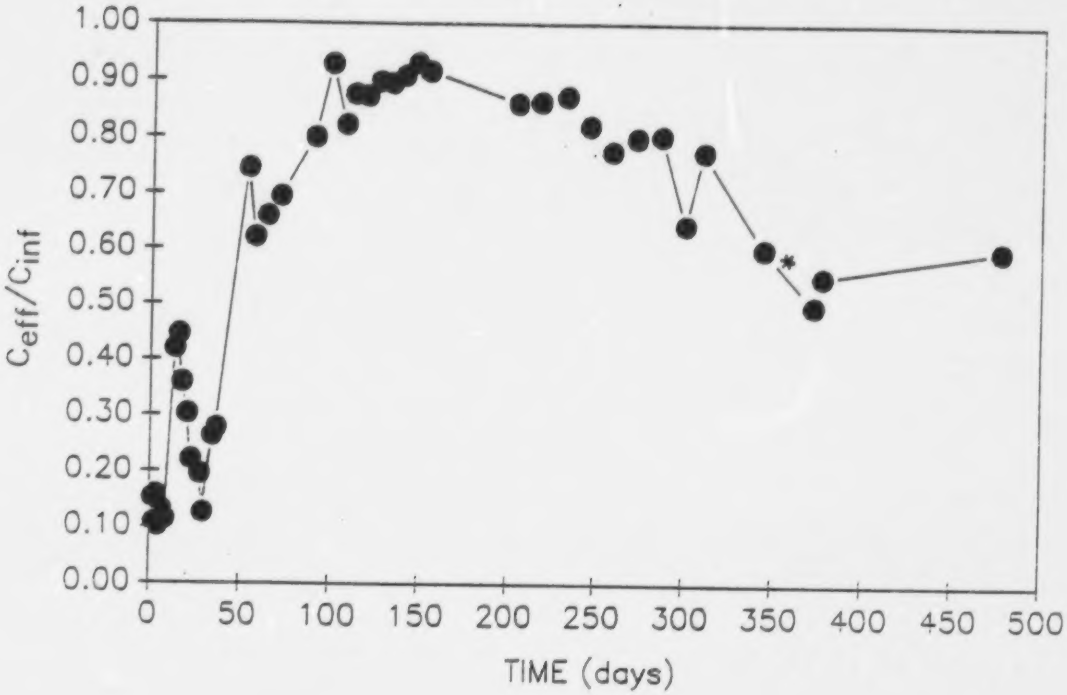


Figure 26. Phase II - Mont Vernon, NH GAC system. Dissolved oxygen. breakthrough curve. (* new well)

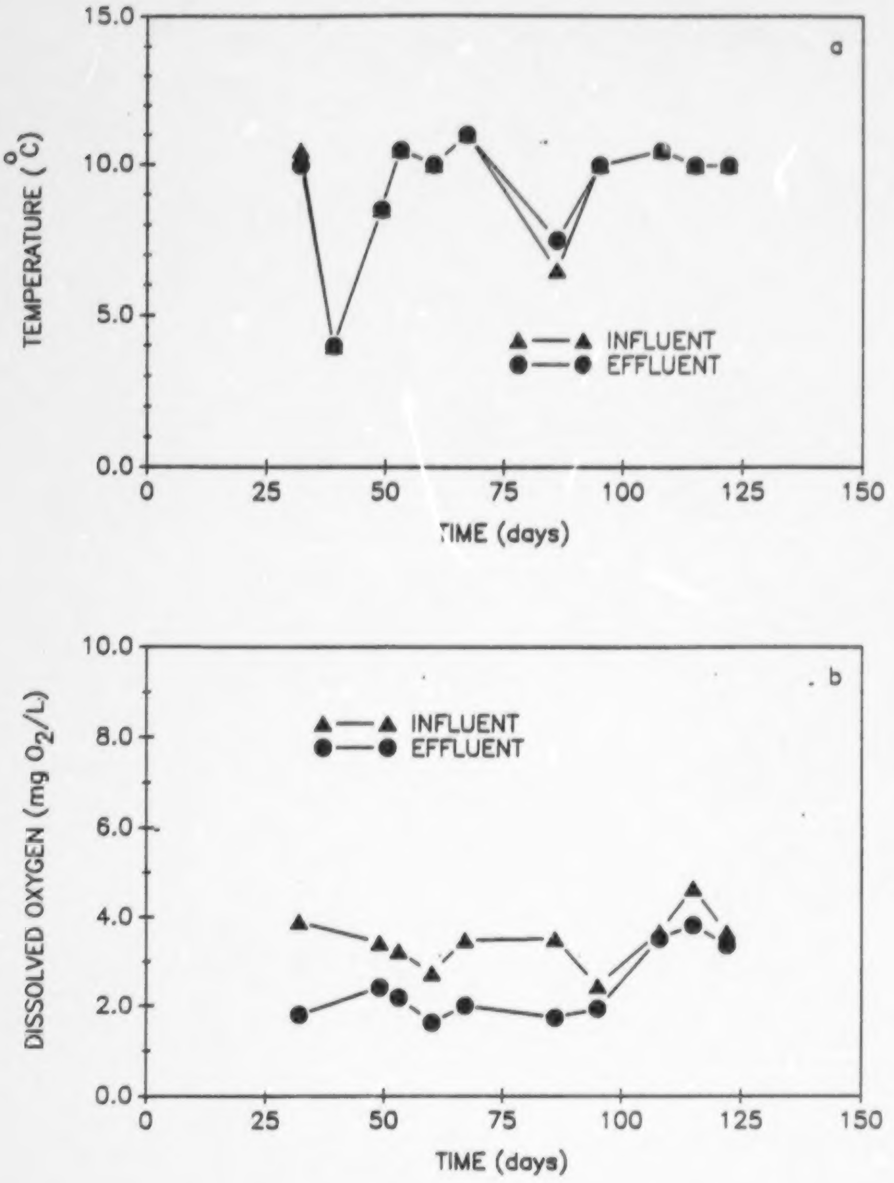


Figure 27. Phase II - Amherst, NH GAC system. Temperature (a) and dissolved oxygen (b) through 122 days of operation.

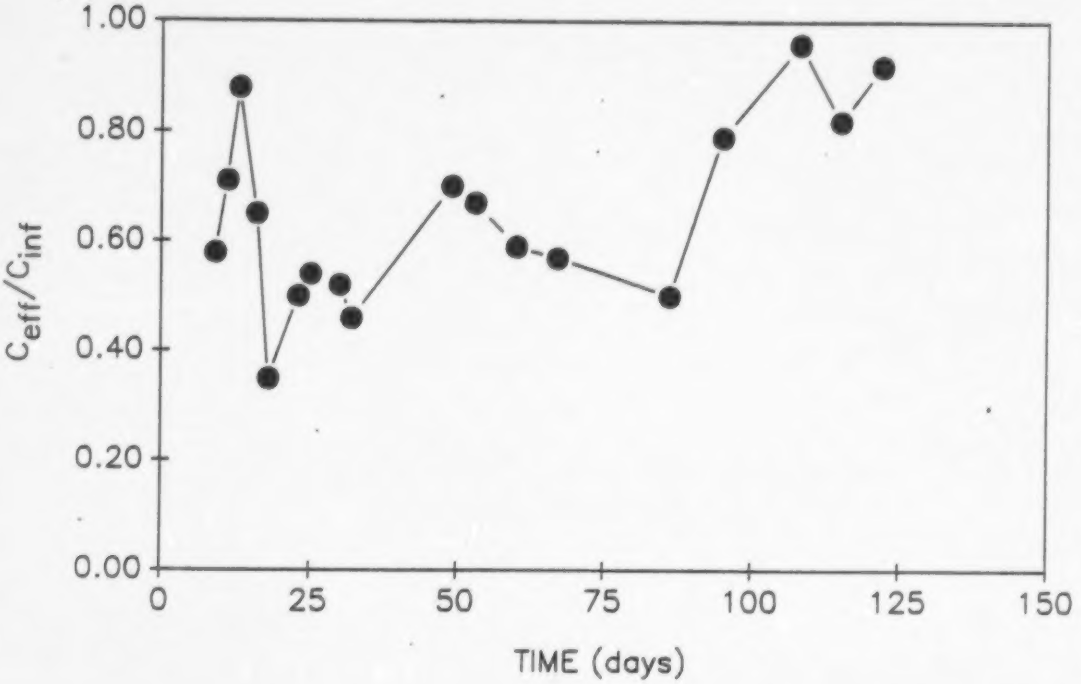


Figure 28. Phase II - Amherst, NH GAC system. Dissolved oxygen breakthrough curve.

- the Amherst unit contained 0.85 m^3 of GAC and was saturated with DO whereas the Mont Vernon unit contained 1.33 m^3 of GAC and continued to adsorb DO.

Turbidity--

Mont Vernon site--As observed during Phase I, turbidity was removed by the GAC units (Figure 29). However, due to the variability in the data and their closeness to the detection limit (0.05 NTU) differences were not significant ($\alpha = 0.05$ and 0.10, StT) between influent and effluent turbidity. Turbidity data taken from the sampling ports indicated there was no distinct trend of removal through the units. Accumulation of small amounts of turbidity may contribute to the potential for clogging the filter, increasing headloss, over long periods of operation. This is particularly important because GAC systems removing radon are not routinely backwashed.

Amherst site--The average influent turbidity (2.99 ± 2.28 NTU) was significantly higher ($\alpha = 0.05$ and 0.01, StT) than the average effluent (1.07 ± 0.50 NTU) (Figure 30). Profiles of turbidity through the system are shown in Figure 31. Turbidity removal was occurring primarily within the top of the GAC unit. The GAC units were not backwashed and thus the accumulation could have been substantial considering a daily flow of 61 m^3/day and an average removal of 1.92 NTU.

Flowrate--

Flowrate data for Phase II were calculated by dividing the total volume of water treated between sampling events by the number of days during that period. As a result, the data only give a gross estimate of the daily flowrate and must be used cautiously, especially during the latter portion of the study when sampling was less frequent.

Mont Vernon site--The average flowrate for the continuous monitoring period was 36 ± 12 m^3/day (Figure 32) which corresponds to an average EBCT of 53.3 min. A dramatic increase in water demand was observed during the period June through August (Day 245 to Day 310), reaching as high as 76 m^3/day (EBCT = 25.3 min). The GAC system was designed to handle an average flowrate of 25 m^3/day (EBCT = 77.9 min). The variability in flow suggests that selecting design flows for small communities using a limited database may be unrepresentative of the true flow variations which may occur. Large variations in flowrate may significantly impact effluent quality because contact time is so critical to GAC adsorption of radon.

Amherst site--The average water flowrate during Phase II was 61 ± 5 m^3/day yielding an average EBCT of 20.1 min and was relatively stable (Figure 33). This flowrate was similar to that observed during Phase I and was much greater than the 35 m^3/day (EBCT = 35.5 min) the system was designed to treat. This could have affected the effluent radon activity because of the shortened contact time within the GAC system. The flow at Amherst was not as variable as Mont Vernon's, but the system was not operated during the summer months when flowrates at Mont Vernon were higher.

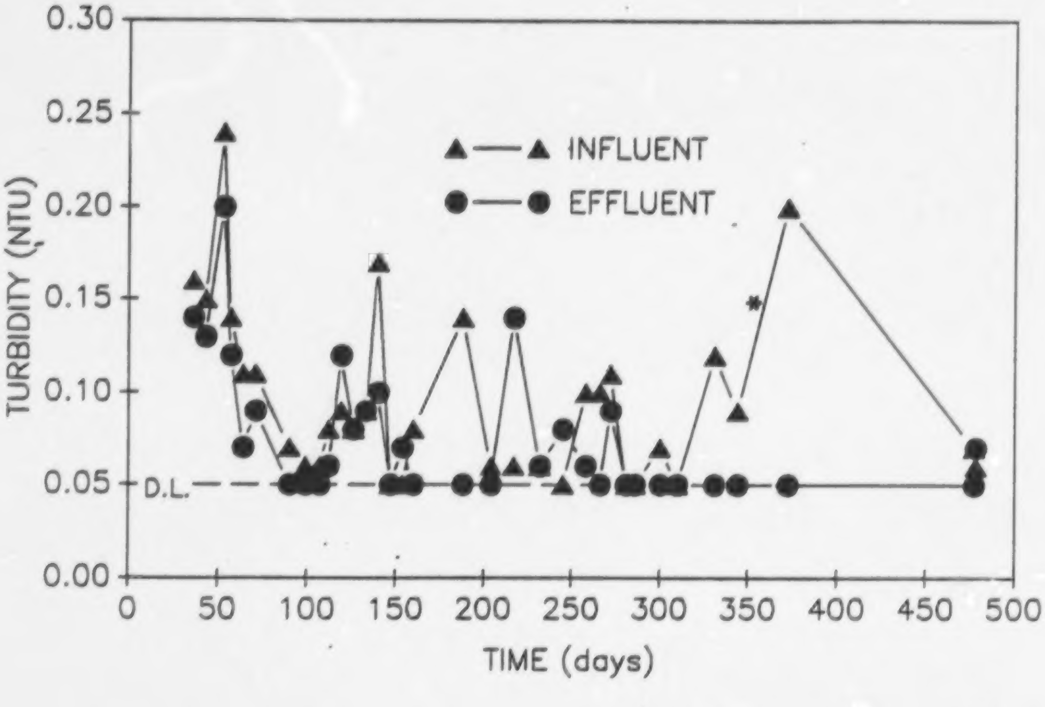


Figure 29. Phase II - Mont Vernon, NH GAC system. Turbidity through 477 days of operation (detection limit = 0.05 NTU). (* new well)

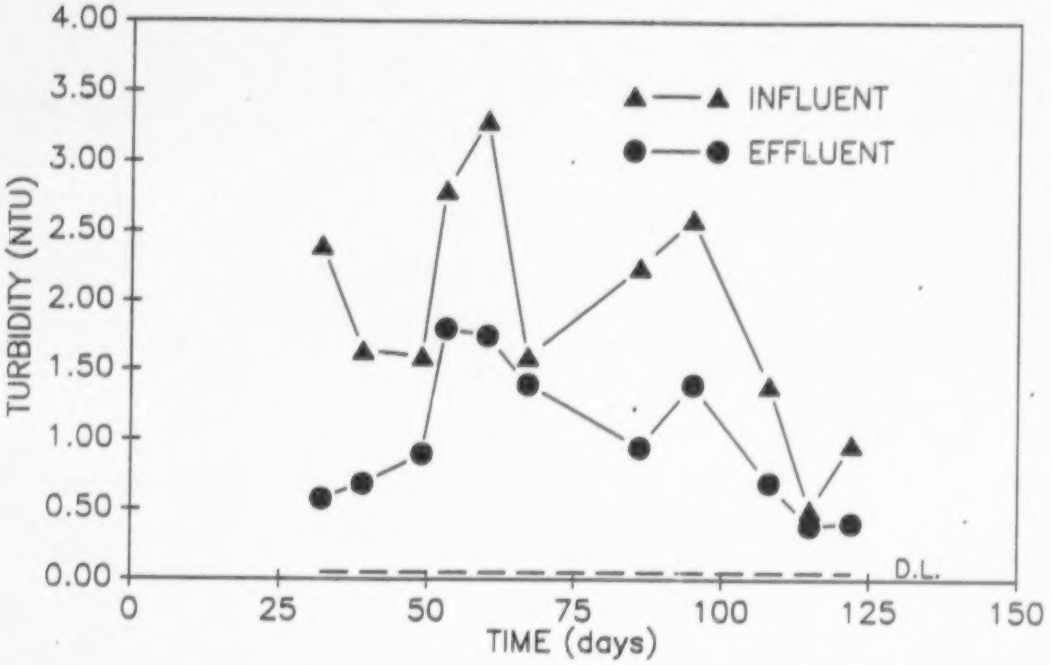


Figure 30. Phase II - Amherst, NH GAC system. Turbidity through 122 days of operation (detection limit = 0.05 NTU).

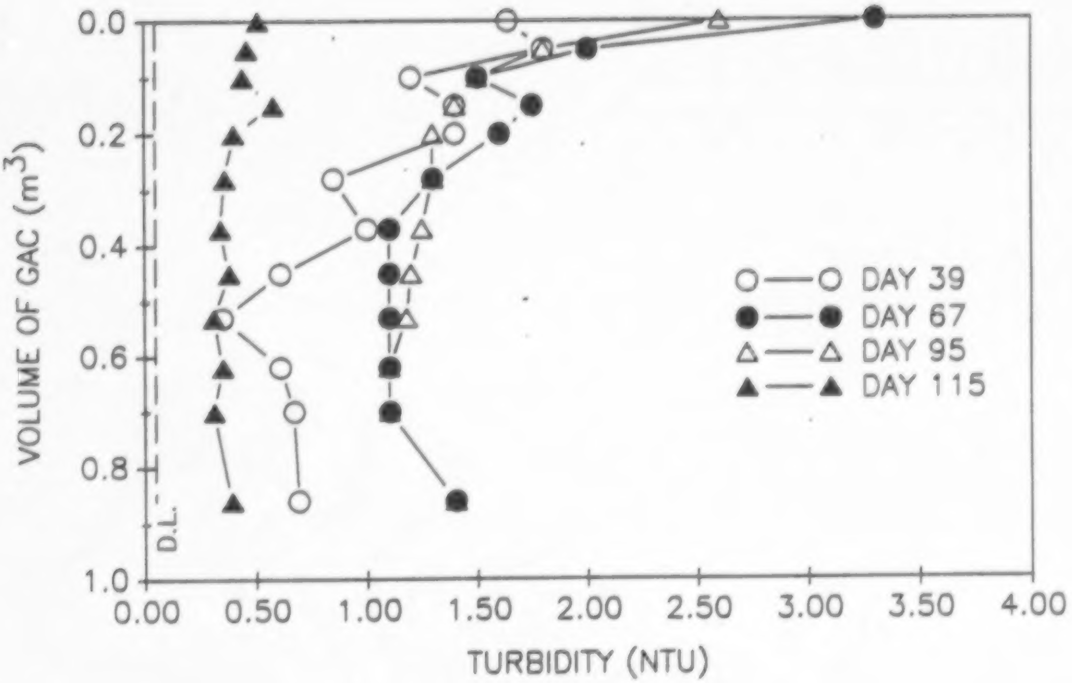


Figure 31. Phase II - Amherst, NH GAC system. Representative profiles of turbidity through the GAC system (detection limit = 0.05 NTU).

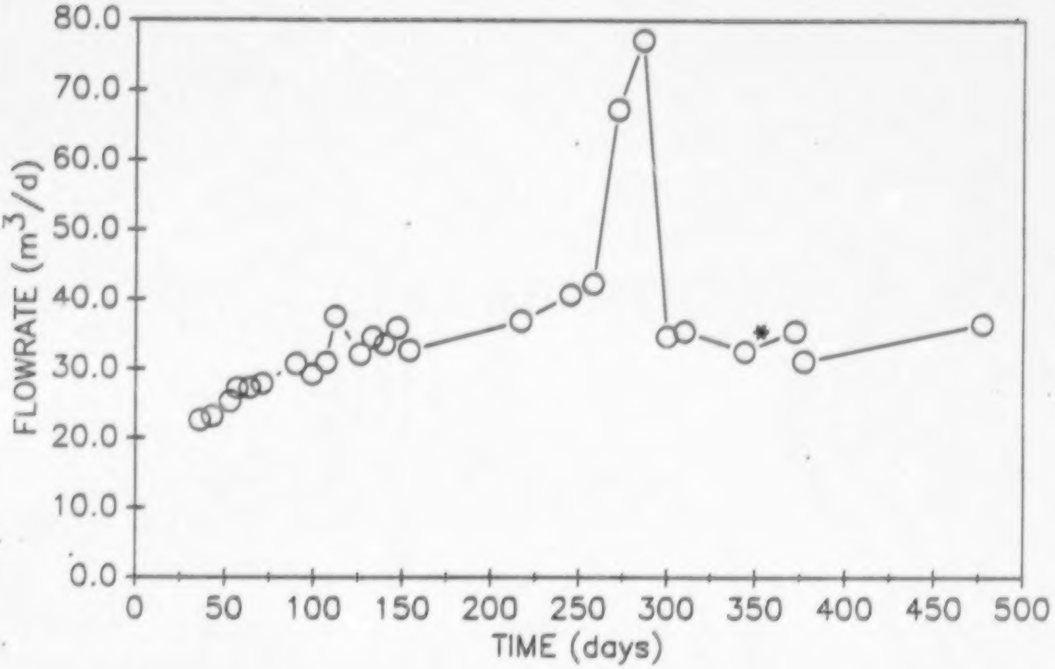


Figure 32. Phase II - Mont Vernon, NH GAC system. Water flowrate through 477 days of operation. (* new well)

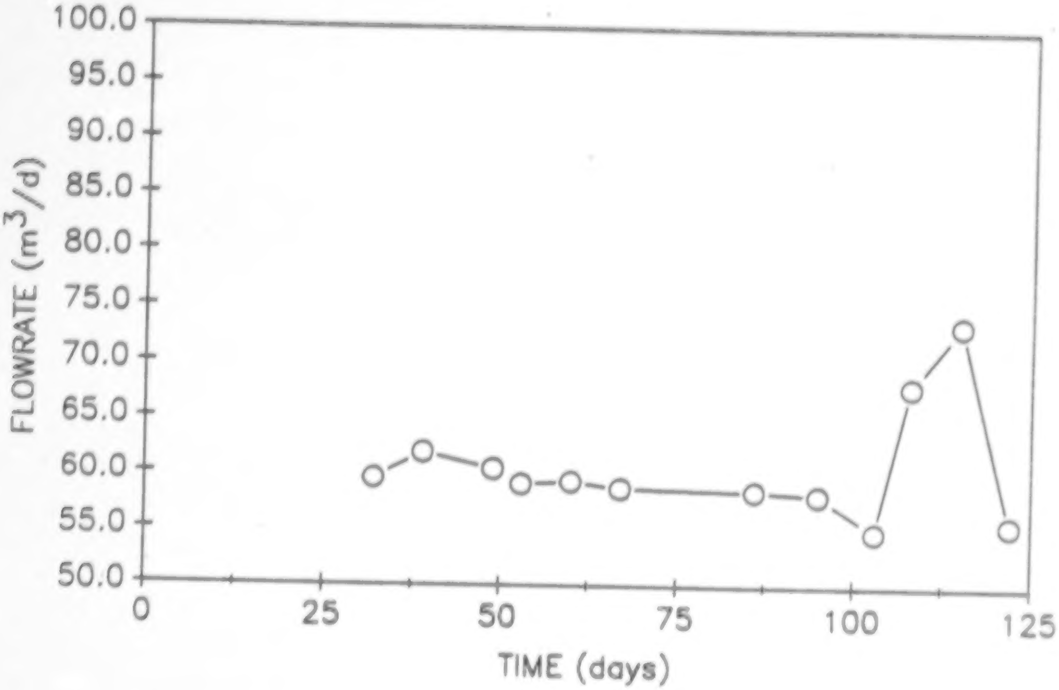


Figure 33. Phase II - Amherst, NH GAC system. Water flowrate through 122 days of operation.

Radon--

Mont Vernon site--With the exception of the 3.5 months at the end of the study when the new well was operating and the average influent activity was 68,900 ± 1,400 pCi/L, the influent radon activity remained higher than the design activity of 155,000 pCi/L. During the majority of Phase II, the average influent activity was 210,491 ± 41,384 pCi/L (Figures 34 and 35). The effluent quality varied between 4,750 to 68,400 pCi/L during Phase II.

Radon is unique among most of the contaminants treated using GAC because after a period of initial breakthrough, there is usually no further increase in the activity observed in the effluent (Lowry and Brandow, 1985). The GAC bed theoretically never becomes exhausted, hence, there is no need for regeneration or replacement. It is believed that this removal pattern occurs because radon decays within the bed.

Profiles of the radon activity through the GAC system are presented in Figure 36. Most of the plots exhibit exponential removal of radon as a function of bed volume which is consistent with the radon adsorption model developed by Lowry (1985). The exception to the pattern occurred late in the study when the new well was operating. Despite the significantly lower influent radon activity from the new well, the data show a very steep (almost straight) plot and an overall decrease in removal efficiency. The Day 477 data is an example of this trend. Although this decrease in removal efficiency corresponds to changes in other water quality parameters, the effects of raw water quality on radon adsorption by GAC are poorly understood.

The K_{ss} of the GAC at Mont Vernon was calculated for periods when flowrate and influent activity were relatively constant. These values averaged 2.37 ± 0.72 hr^{-1} as compared to the K_{ss} of 3.02 hr^{-1} at 6-10°C obtained in other tests with this type of GAC (Lowry and Lowry, 1987). Differences in the quality of the water treated by these systems and the column hydraulics may have accounted for these differences in K_{ss}.

Amherst site--The average influent radon activity during Phase II was 41,800 ± 15,200 pCi/L (Figure 37), which was not significantly different (α = 0.05 and 0.01, StT) from the design influent of 39,730 pCi/L. The influent radon activity was lower during this period than during Phase I and also showed a decreasing trend with time. The average effluent radon activity observed was 12,000 ± 5,900 pCi/L. Figure 38 presents profiles of radon activity through the Amherst GAC system. The activities observed in the section of the bed between 0.53 and 0.70 m^3 were sometimes lower than the effluent activity probably because air was entrained in the port tubing during sampling.

Again, the predicted (Lowry, 1985) exponential radon removal pattern was observed. K_{ss} values through the GAC unit average 5.7 ± 1.04 hr^{-1} during Phase II, which were considerably higher than the design K_{ss} of 3.02 hr^{-1} at 6-10°C previously obtained in other studies with this GAC (Lowry and Lowry,

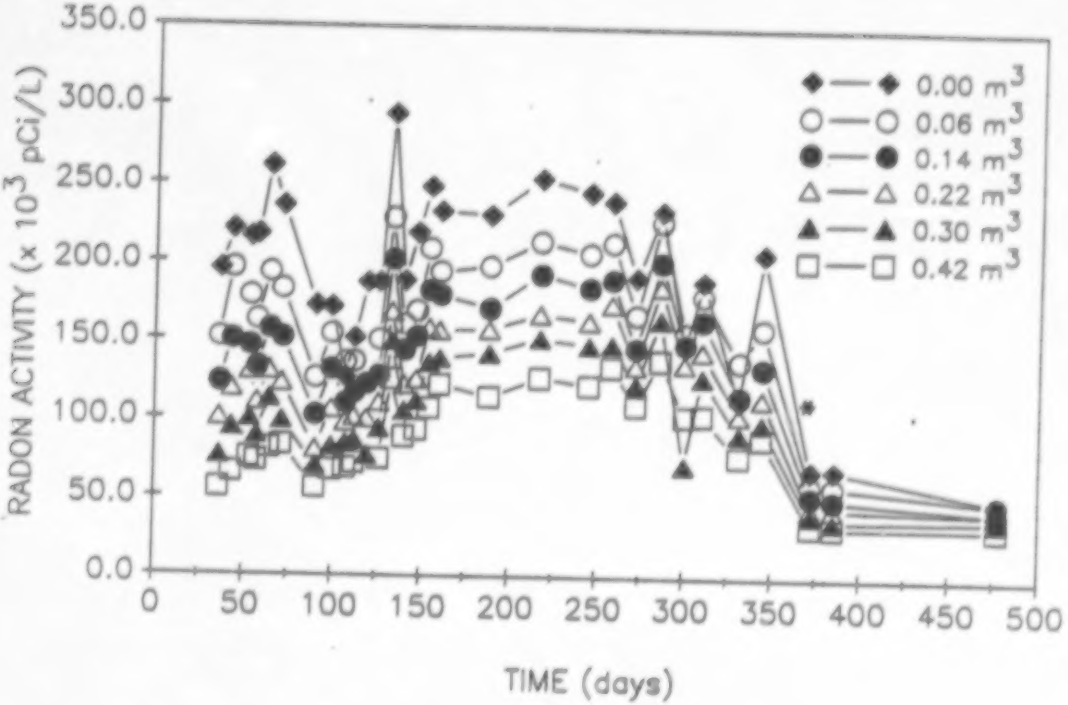


Figure 34. Phase II - Mont Vernon, NH GAC system. Radon activity through GAC #1 through 477 days of operation. (* new well)

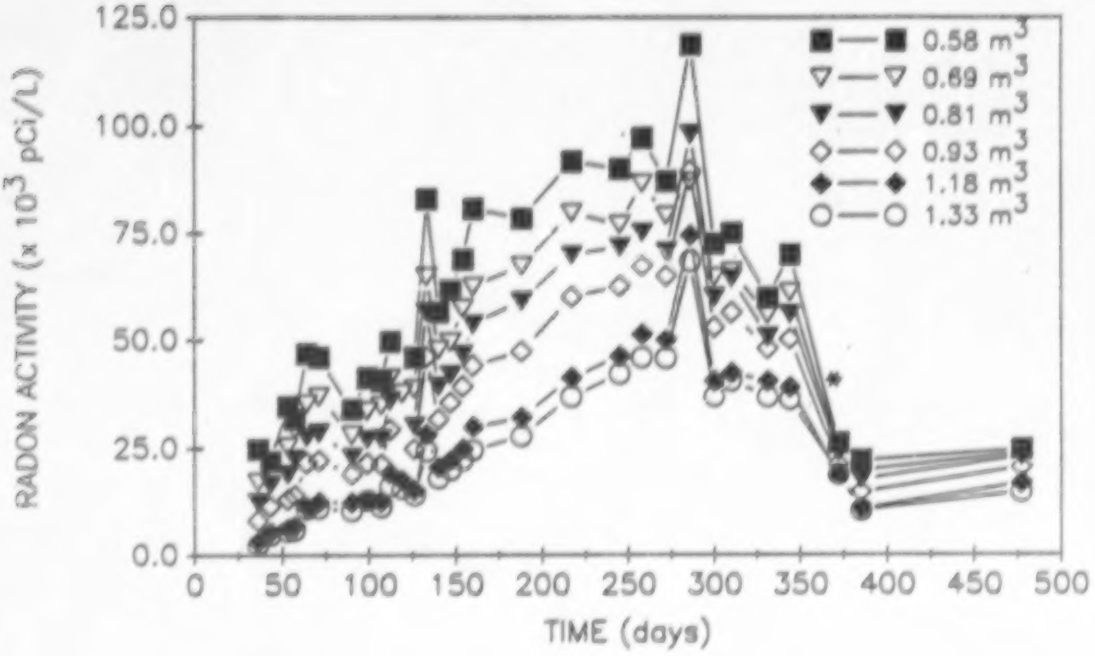


Figure 35. Phase II - Mont Vernon, NH GAC system. Radon activity through GAC #2 through 477 days operation. Note scale difference between Figure 34 and 35. (* new well)

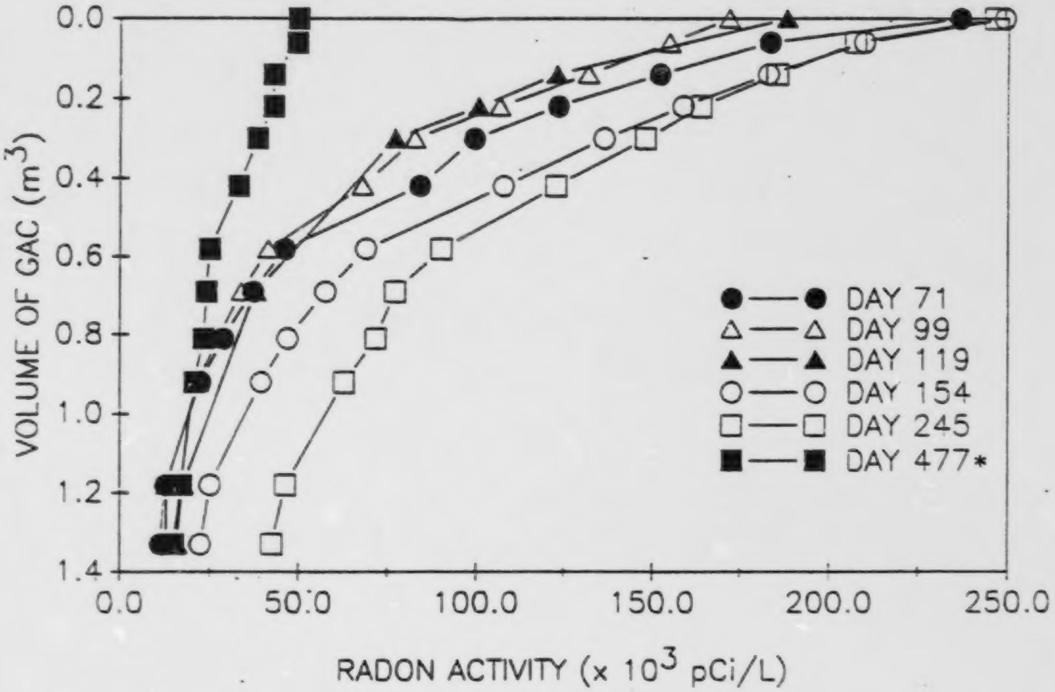


Figure 36. Phase II - Mont Vernon, NH GAC system. Representative profiles of radon activity through the GAC system. (* new well)

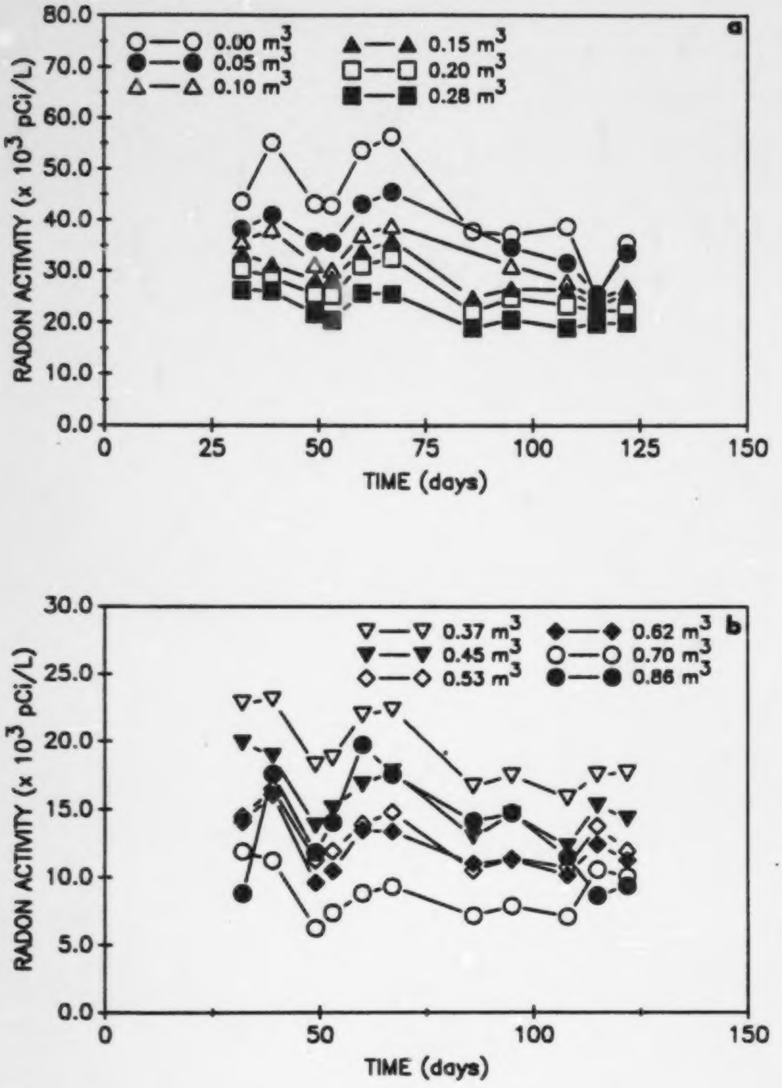


Figure 37. Phase II - Amherst, NH GAC system. Radon activity through the top 0.28 m³ of GAC (a) and the remaining 0.58 m³ of GAC (b) during 122 days of operation. Note difference in scales on ordinates in (a) and (b).

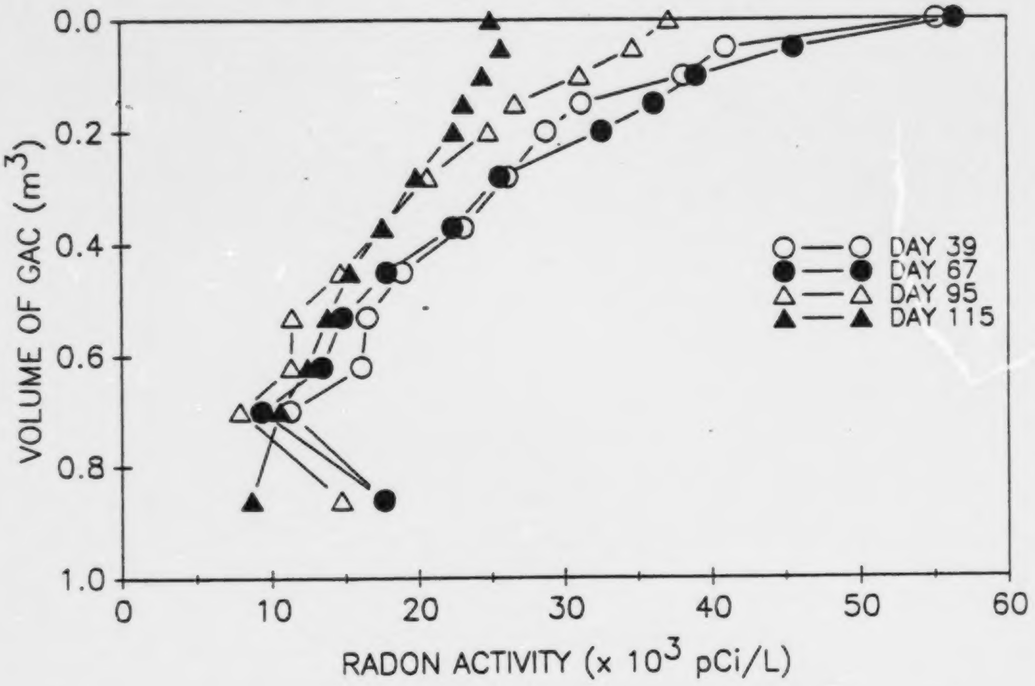


Figure 38. Phase II - Amherst, NH GAC system. Representative profiles of radon activity through the GAC system.

1987). The one exception was the K_{ss} data obtained during stable flow and radon activities in the GAC from 0.53 to 0.70 m^3. The K_{ss} in that region averaged 0.55 to 0.32 hr^{-1}, but may reflect problems with air entrainment in the sampling tubing.

One of the problems with designing GAC systems for radon removal is the need for data on influent radon activity and flowrate. These two parameters can vary widely in small community water supplies as demonstrated in this study. Therefore, design could be difficult unless a community had routinely monitored water quality and flowrate. Though the data shows the GAC systems can accommodate some variation in influent loading, changes in flow and/or radon activity typical in small community use of groundwater supplies could result in spikes of radon in the effluent exceeding the MCL. Further, since GAC system design is based on the K_{ss} (Lowry et al., 1987; Lowry and Lowry, 1987) and in our evaluations at Mont Vernon and Amherst this value was found to vary depending on site conditions (i.e., flowrate, radon concentration and raw water quality), it is recommended that pilot scale testing be performed to verify the GAC's capacity for radon at a given site prior to finalizing the system's design.

Gamma/Beta Emissions--

Mont Vernon site--Gamma/beta emissions in the top and middle of GAC #1 began to converge and were not significantly different (α = 0.05 and 0.01, StT) after 225 days of operation (Figure 39). Gamma readings for GAC #2 resembled the fluctuations observed for the radon activities in that unit. As observed during Phase I, there was a curvilinear relationships between influent radon activity and gamma/beta emissions readings (Figure 40). After 6 months of operation and before the new well was used, the relationships between gamma/beta emissions and the influent radon activity at the top of GAC #1 and #2 were 1 mR/hr = 5,570 pCi/L and 1 mR/hr = 6,549 pCi/L, respectively. These relationships are different than those observed by Lowry et al. (1988) (1 mR/hr ≡ 10,360 pCi/L) for point-of-entry units. The fluctuations in radon load (radon activity and flow) and removal could have had a tremendous effect on the gamma emissions through the Mont Vernon GAC units and the large GAC volume could account for some of the differences observed between the small community systems and the point-of-entry units. Perhaps, a mass balance approach would provide a better mechanism to compare emissions from different systems since it accounts for differences in the mass of GAC and radon mass loading. However, there is still a problem with geometries of the source and the differences in monitoring instrumentation which may make comparisons between GAC systems difficult. This may be less of a problem when comparing units with relatively similar flows, GAC masses, and contactor geometries.

Keene and Rydell (1989) cite a maximum 8 hr/day exposure applicable to residences of 0.058 mR/hr based on National Council on Radiation Protection guidelines. The data obtained at Mont Vernon indicates that gamma exposure in the 10^1 mR/hr range could occur at the units' surfaces. The background gamma/beta emissions measured in an area approximately 31 m away from the

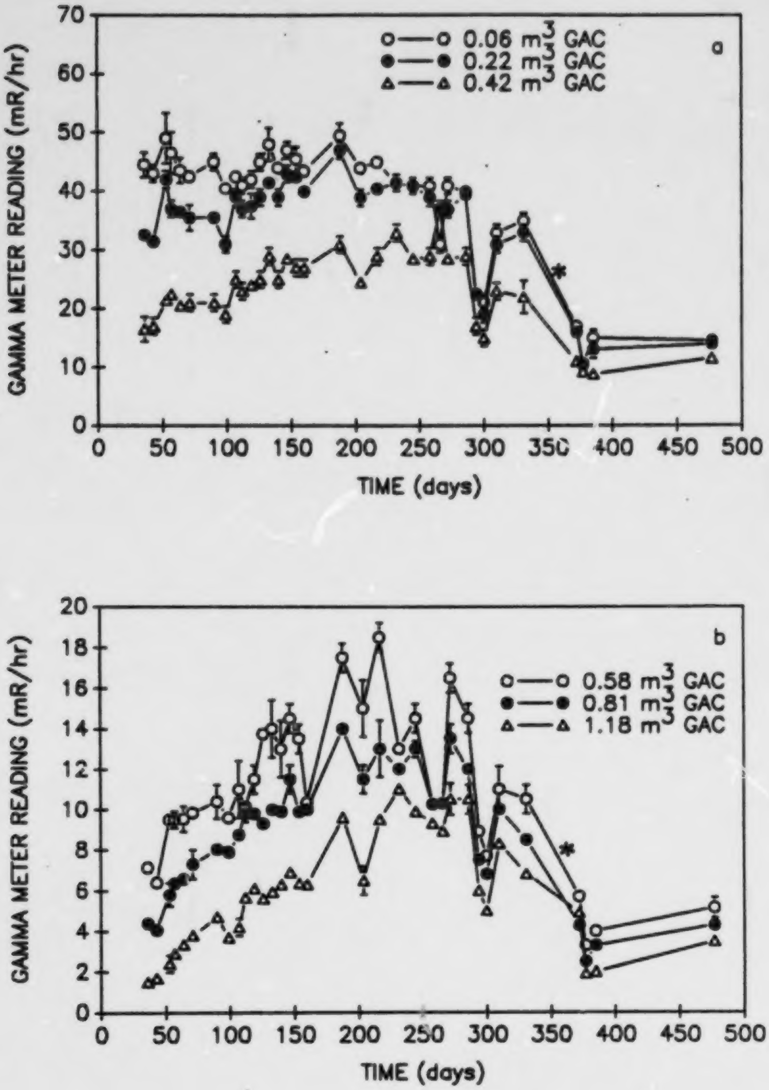


Figure 39. Phase II - Mont Vernon, NH GAC system. Gamma/beta emission measurements taken on the contactor surfaces through 477 days of operation for GAC #1 (a) and GAC #2 (b). Note difference in scales on ordinate in (a) and (b). (* new well)

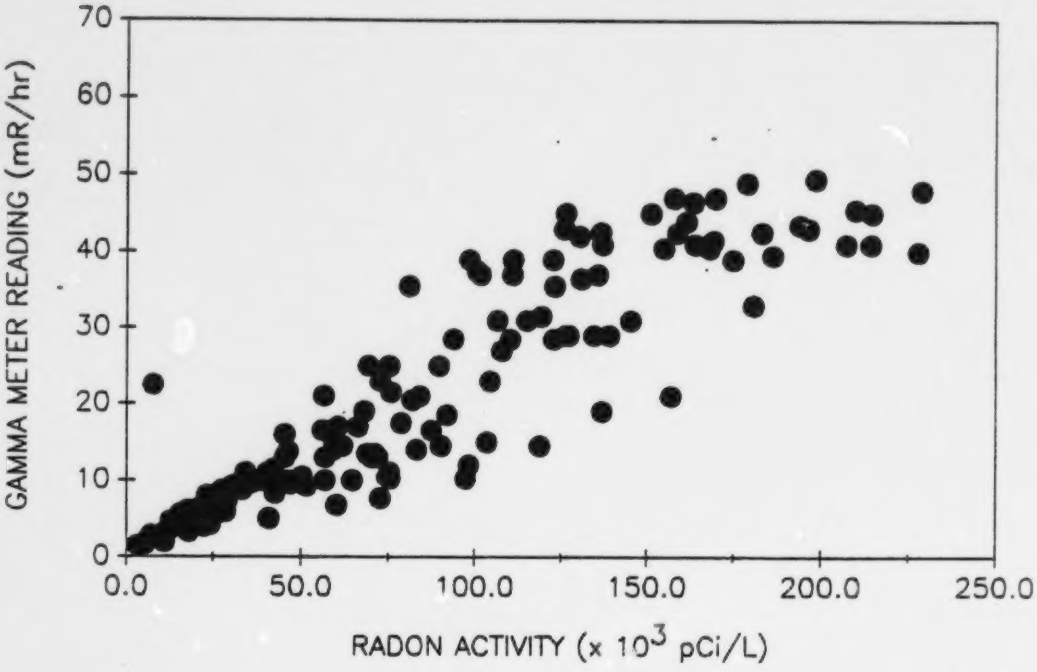


Figure 40. Phase II - Mont Vernon, NH GAC system. Gamma/heta emission measurements taken on the contactor surfaces as a function of effluent radon activity to sections of the GAC system.

pumphouse averaged 0.056 ± 0.028 mR/hr. The gamma/beta emissions measured in the vicinity of the GAC system decreased with distance from the units. 1.52 m away from GAC #1 the gamma/beta readings average 3.56 ± 0.97 mR/hr (same elevation as the middle of the tank), while 0.91 m from GAC #2 the readings averaged 1.2 ± 0.65 mR/hr (same elevation as middle of the tank).

To corroborate that gamma exposures were significant at the Mont Vernon site, dosimeter badges were placed in the pumphouse and on the surface of the GAC for 4 days. This was done when the influent flowrate and radon activity to the units were approximately 50 m^3/day and 53,304 pCi/L, respectively. The badge on the middle of GAC #1 measured 970 mREM, while a badge at the same elevation 0.61 m away from the unit registered 290 mREM. The badge on the middle of GAC #2 measured 400 mREM, while a badge 0.61 m away measured 160 mREM.

Exposures are highly dependent on shielding and the distance away from the source. Gamma exposures from small community GAC systems should be predicted based on radon removal (flowrate, influent radon activity, effluent quality, GAC volume and tank configuration) and should be minimized to meet accepted occupational safety and health standards.

Amherst site--The gamma/beta emissions during Phase II decreased gradually over the period as observed for the radon activities (Figure 41). Figure 42 shows the relationship between gamma/beta emissions and the radon activity at the nearest port. The non-linear relationship is similar to the one observed during Phase I for this system. Again, these differences may be a function of the mass of GAC, the radon loading/removal, the tank geometry and/or the instrumentation used. The background gamma/beta emissions at Amherst, approximately 9 m from the pumphouse, were 0.027 ± 0.031 mR/hr. Emissions in the doorway, approximately 1.2 m above the filter, averaged 0.33 ± 0.15 mR/hr.

Microbial Numbers--

Mont Vernon site--It has been well documented that GAC filters are capable of supporting microbial populations (Wilcox et al., 1983; Camper et al., 1985, 1986, 1987). Effluent standard plate count (SPC) microbial enumerations are often greater than influent SPC for GAC systems used in water treatment (Graese et al., 1987). The GAC provides a good surface for attachment and concentrates nutrients to support the microorganisms. The microbial numbers in the GAC effluent fluctuated greatly after 125 days of operation, but were significantly higher ($\alpha = 0.05$ and 0.01, StT) than the influent concentration (Figure 43).

Amherst site--The microbial numbers in the effluent from the GAC at Amherst were highly variable (Figure 44), as observed for the Mont Vernon site. There was some input of microorganisms to the unit from Day 53 to 67. It appears that the GAC at Amherst also supported a microbial population.

The microbial data collected for the GAC units at Mont Vernon and Amherst show significantly different conditions existed at each site. The Mont Vernon data indicates bacterial populations in the GAC effluent were significantly

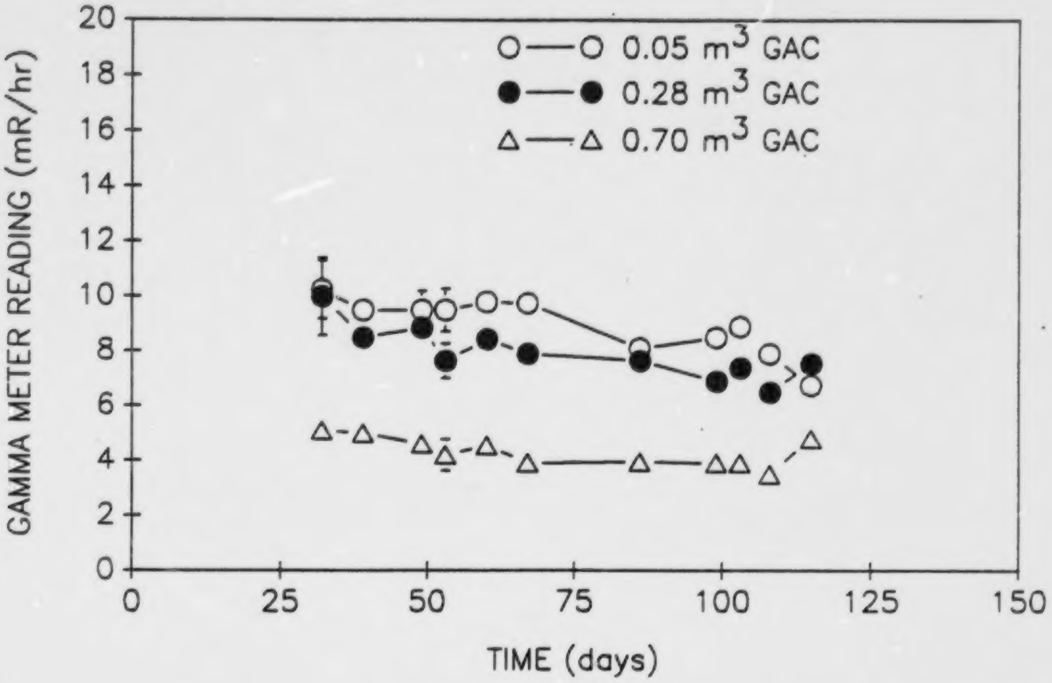


Figure 41. Phase II - Amherst, NH GAC system. Gamma/beta emission measurements taken on the contactor surface through 122 days of operation.

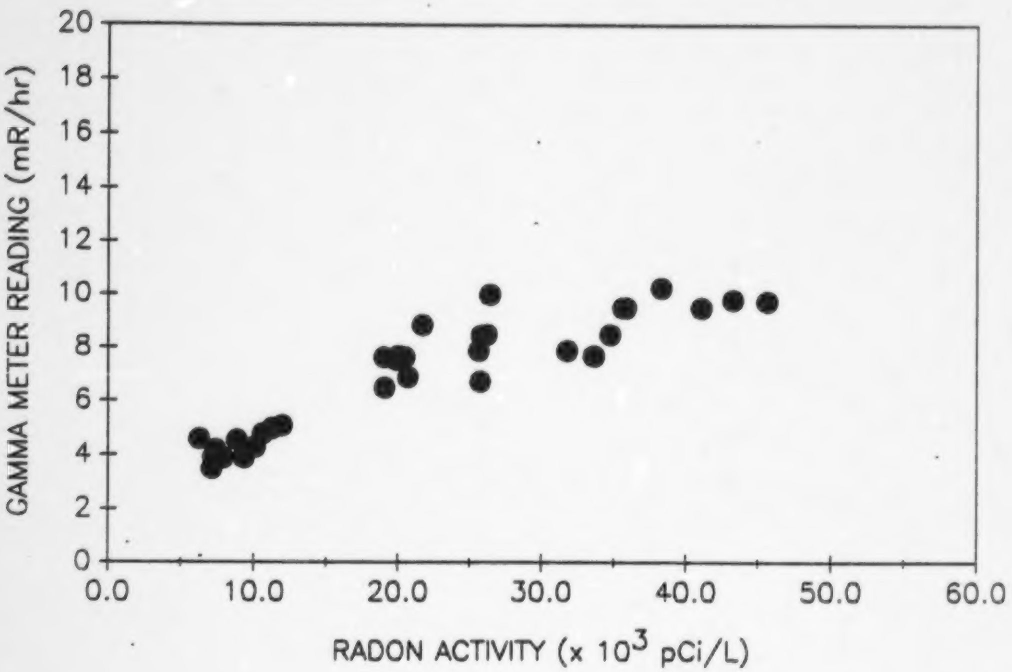


Figure 42. Phase II - Amherst, NH GAC system. Gamma/beta emission measurements as taken on contactor surface as a function of influent radon activity to sections of the GAC system.

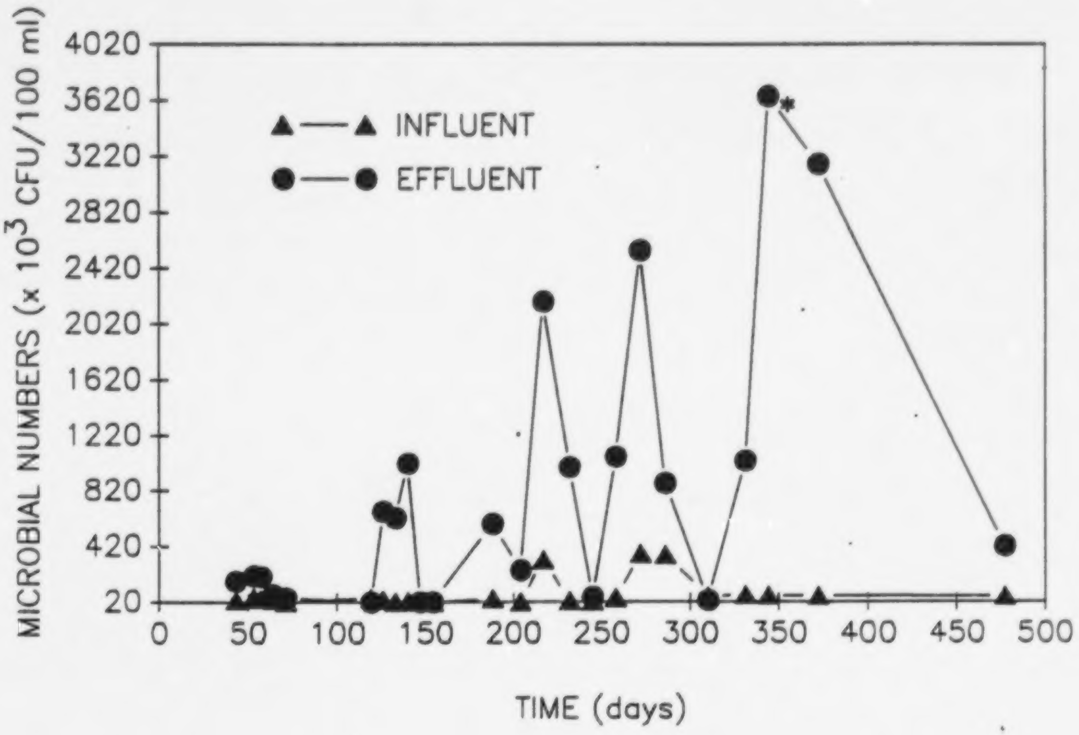


Figure 43. Phase II - Mont Vernon, NH GAC system. Microbial numbers through 477 days of operation (detection limit = 20,000 CFU/100 mL). (* new well)

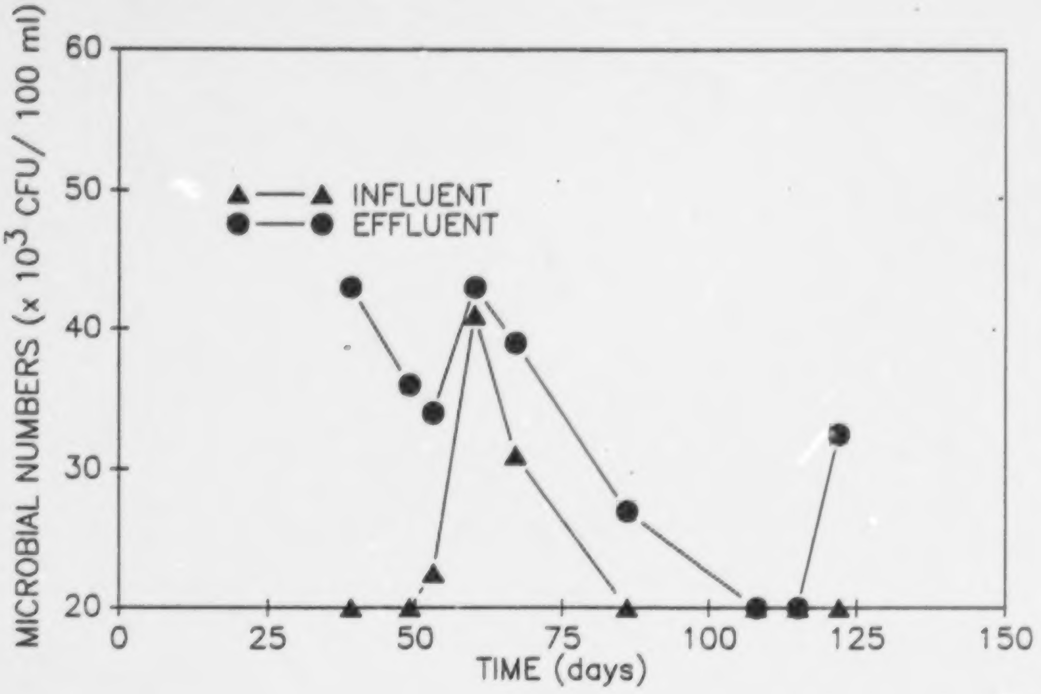


Figure 44. Phase II - Amherst, NH GAC system. Microbial numbers through 122 days of operation (detection limit = 20,000 CFU/100 mL).

higher than the typical ranges of 0 to 3,000 colonies/mL reported in the literature (Wilcox et al., 1983) for GAC columns used in water treatment. It is important to note that the bacterial concentrations found in the influent at Mont Vernon were elevated. Results for Amherst indicated that the microbial numbers for the GAC effluent ranged from about 200 to 400 colonies/mL. These values fall within the range of 0 to 600 colonies/mL reported by Bourbigot (1981) in effluent from a GAC column. The variations and fluctuation observed in the microbial number are analogous to those observed by other researchers (Wilcox et al., 1983) and may be related to seasonal effects.

Iron and Manganese--

Mont Vernon site--The iron and manganese concentrations during Phase II were mostly at or below the detection limit (Fe = 0.06 mg/L; Mn = 0.02 mg/L). On Day 43, the influent manganese concentration was 1.0 mg/L. Manganese was never detected in the GAC effluent. Iron concentrations were below detection limit in the effluent. Though the overall change in concentration, if it occurred, was probably small, the total accumulation of these metals in the GAC units may have been substantial considering the flow treated. For example, assuming an average flow of 36 m^3/day and a removal of 0.02 mg Fe/L, 262 g of Fe would have accumulated over a year. This may cause a problem in the units after several years of operation considering that backwashing is not normally recommended for GAC systems removing radon.

Amherst site--The water in Amherst contained significantly higher (α = 0.05 and 0.01, StT) concentrations of iron (0.78 ± 0.98 mg/L) than at Mont Vernon (Figure 45). Manganese concentrations averaged 0.10 ± 0.21 mg/L during Phase II and were significantly higher than Mont Vernon's at $\alpha = 0.10$, but not $\alpha = 0.05$ (StT). The average effluent iron and manganese concentrations were 0.16 ± 0.14 mg Fe/L and 0.08 ± 0.16 mg Mn/L, respectively. Figures 46 shows profile data for iron through the Amherst GAC unit. Iron was removed (influent vs. effluent iron significantly different at $\alpha = 0.05$, but not at 0.02; StT) on some days in the upper portion of the filter. There was no significant change (α = 0.05 and 0.10, StT) in manganese through the filter.

Uranium and Radium--

Mont Vernon site--The average uranium activity of the influent water from the original wells was 24.6 ± 3.6 pCi/L. Uranium removal followed the theoretical profile of a well adsorbed solute (Figure 47). Removal was not similar to radon because uranium species have very long half-lives (~10^5 to 10^9 yr). As a result, decay did not significantly impact the fate of sorbed uranium over the course of this study. On Day 43, all of the uranium (detection limit = 0.1 pCi/L) was removed after passing through 0.57 m^3 of GAC. Most of the uranium was removed by the top 0.57 m^3 of GAC on Day 300, with some removal occurring in the remaining 0.76 m^3. The removal profile over time indicated that the GAC bed would eventually be exhausted with respect to uranium.

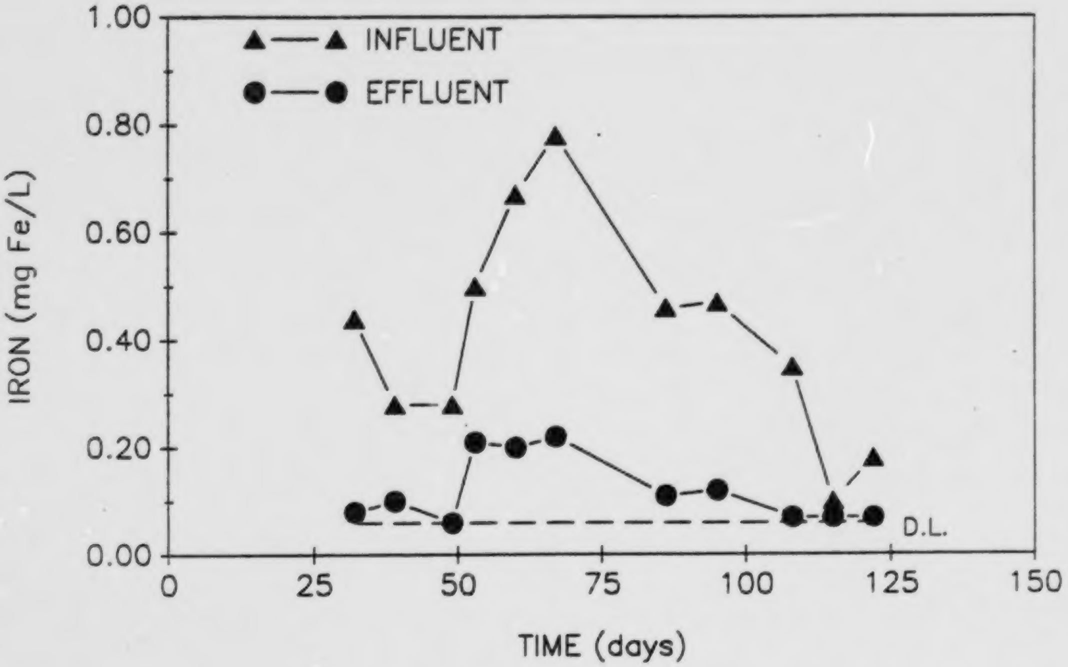


Figure 45. Phase II - Amherst, NH GAC system. Iron data through 122 days of operation (detection limit = 0.06 mg/L).

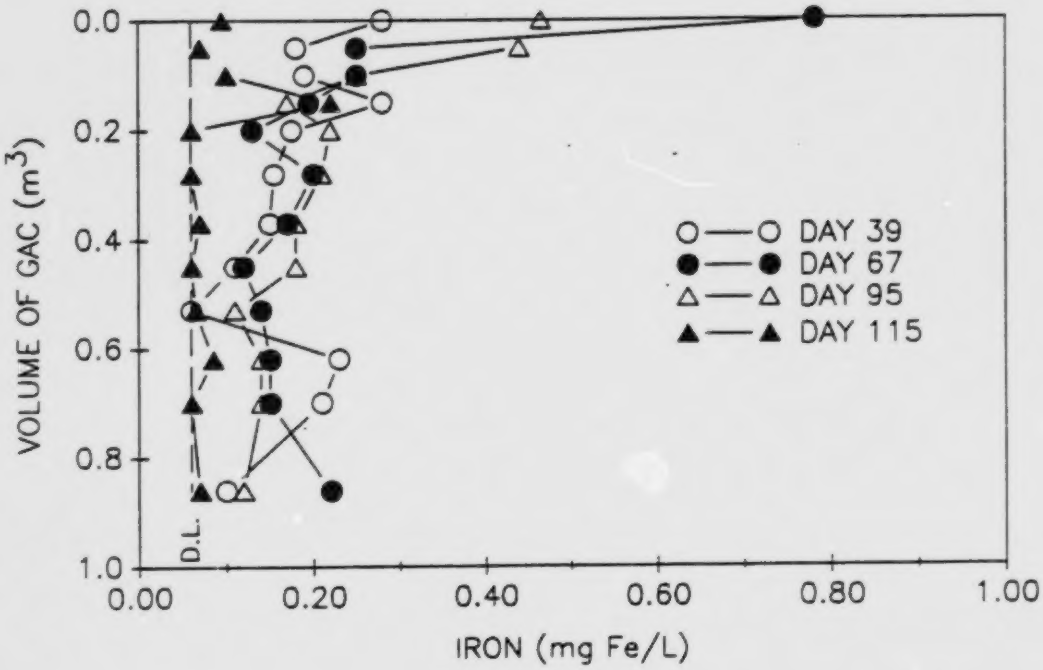


Figure 46. Phase II - Amherst, NH GAC system. Representative profiles of iron through the GAC system (detection limit = 0.06 mg/L).

The uranium activity increased dramatically (>500 pCi/L) when the new well was installed. Figure 47 shows the data collected during this period. By Day 372, the system had begun experiencing fluctuations in the uranium adsorption profile through the units. By Day 477, the influent uranium activity approached 500 pCi/L, however, there was little uranium removal.

The change in uranium removal exhibited by the GAC may have been due to a shift in uranium speciation resulting from the change in pH and alkalinity of the new water source. The effective removal of uranium by anion exchange (Jelinek and Sorg, 1988) suggests that uranium removal by GAC is based on coulombic (ionic or electrical) adsorption. Since researchers (Bean et al., 1964) have demonstrated that the surface of GAC is primarily nonpolar with a net negative charge, it follows that adsorption is at a minimum for negatively charged species and at a maximum for neutral species (Weber, 1972) assuming highly ionized positive species tend to be hydrophillic and therefore also reduce adsorption. Relating these facts to uranium speciation, Sorg (1988) reported that above pH 7 (new well = 7.7 ± 0.03) the negatively charged uranyl carbonate complex ($UO_2(CO_3)_2^{2-}$) is the predominant species whereas below pH ~ 6.8 (old well = 6.5 ± 0.2) the neutral UO_2CO_3 species predominates. Therefore, it is hypothesized that the shift in raw water pH and alkalinity when the new well was installed at Mont Vernon resulted in a shift in uranyl carbonate distribution from the favorably adsorbed neutral UO_2CO_3 to the poorly adsorbed $UO_2(CO_3)_2^{2-}$ species.

The radium in the water supply increased from 1.4 ± 0.1 pCi/L to >19 pCi/L when the new well was used (Figure 48). Data for influent and effluent radium were only available for a limited number of days. On Day 245, the effluent radium activity equaled the influent activity. When the new well began operating, the influent of 19.3 pCi/L on Day 372 was reduced to 2.3 pCi/L in the effluent. By Day 477, only 1.8 pCi/L of radium was removed by the GAC system (influent = 18.4 pCi/L, effluent = 16.6 pCi/L).

It is well documented that radium is poorly adsorbed by GAC (Sorg and Logsdon, 1978) since it has the greatest ionic and electropositive nature of all the Group II elements (Cotton and Wilkinson, 1980) rendering it extremely hydrophillic. Therefore, the small changes in radium concentration observed are not surprising. The observed removal of radium on Day 372 may be a result of physical filtration of radium containing particles by the GAC bed (Faust and Aly, 1987) or adsorption or ion exchange between the radium and precipitated solid phases (e.g., $Fe(OH)_3$) and/or organic matter deposited within the GAC bed.

Presently, there is no MCL set for uranium, but it is likely to be in the range of 20 to 40 pCi/L (EPA, 1989). The MCL for total radium is 5 pCi/L. Both uranium and radium present serious health concerns. Though GAC has some capacity for adsorbing these radionuclides, it seems to be sensitive to pH and alkalinity and it is not an alternative treatment technique for these radionuclides. In the Mont Vernon system, additional treatment for these radionuclides would be required to meet the MCLs. Adsorption of uranium and radium to the GAC may also be problematic because of their long half-lives.

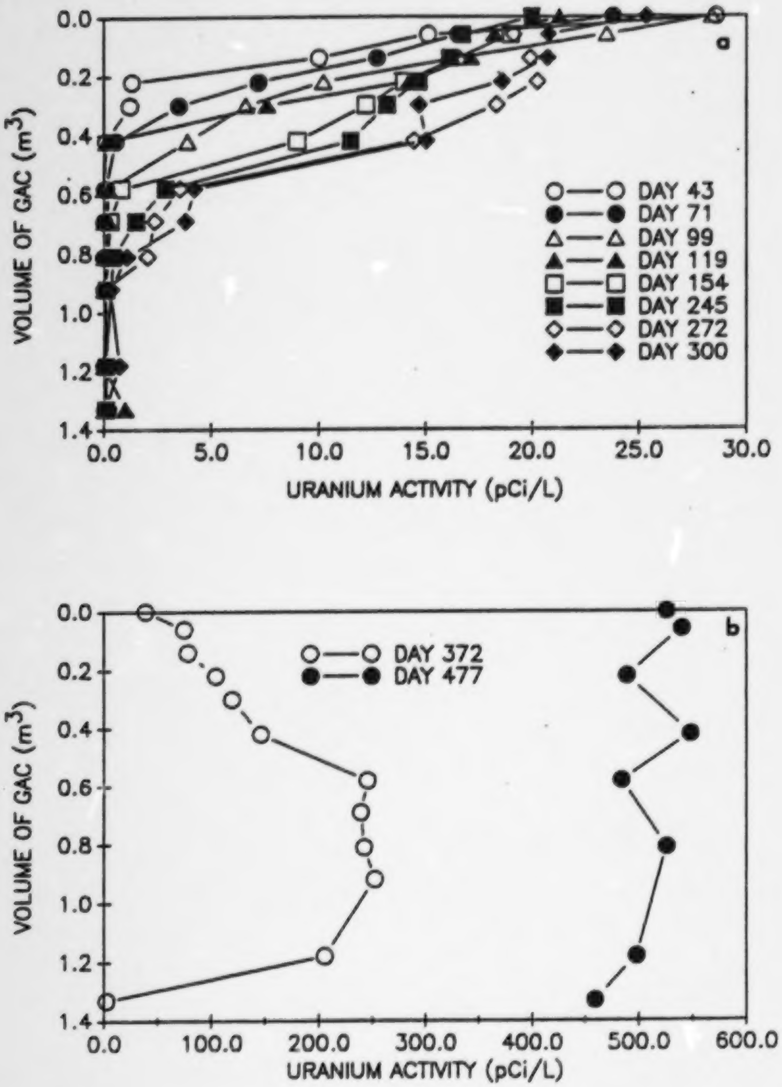


Figure 47. Phase II - Mont Vernon, NH GAC system. Representative profiles of uranium through the GAC system. Note differences in scales on abscissa. Original well (a) and new well (b).

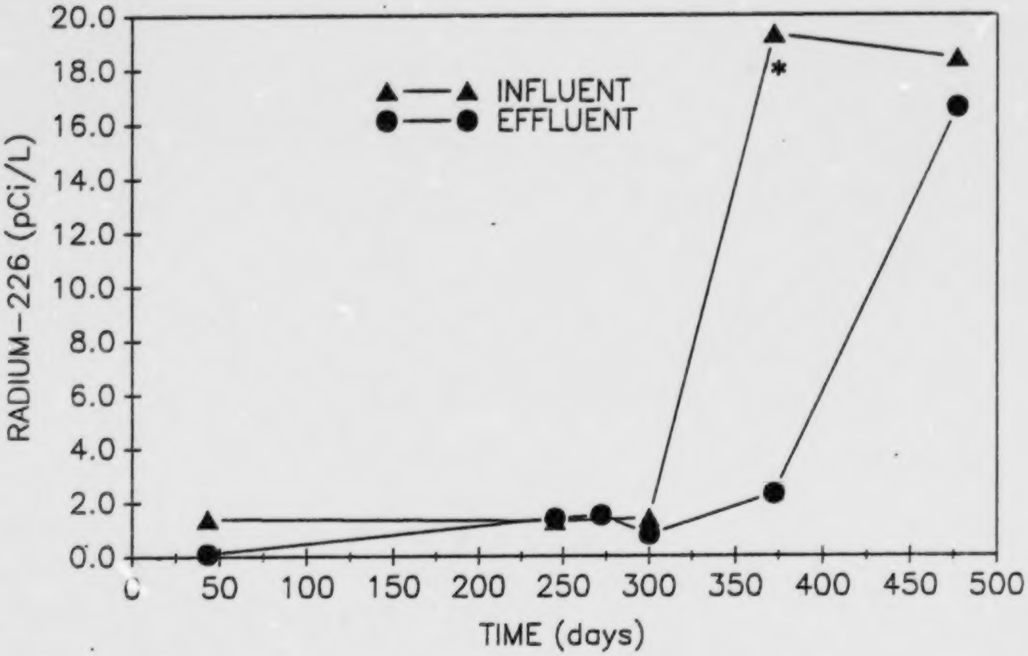


Figure 48. Phase II - Mont Vernon, NH GAC system. Radium-226 data through 477 days of operation. (* new well)

Accumulation of these radionuclides could lead to disposal problems with the GAC.

Amherst site--The uranium in the Amherst water supply averaged 34.5 ± 20.2 pCi/L. There was a steady decrease in the influent uranium activity during Phase II. The typical exponential removal profiles observed at Mont Vernon for uranium were not found at Amherst except during Day 39 (Figure 49). On many instances, higher uranium activities were observed within the unit than in the influent water. The pH and alkalinity of the Amherst water averaged 8.0 ± 0.14 and 48 ± 4.3 mg/L as $CaCO_3$, respectively during Phase II. As previously discussed, under these conditions, the negatively charged $UO_2(CO_3)_2^{2-}$ species will predominate and tend not to sorb to the GAC.

The only radium data available at Amherst was for Day 39 where the influent activity of 2.1 pCi/L was reduced to an effluent of 0.7 pCi/L. The limited database for radium prevented analysis of radium removal by the GAC system at Amherst.

Special Sampling Events

Diurnal Variations--

The effect of diurnal variations was investigated at the Mont Vernon site for a period of 12 hr on two consecutive days in July, 1987 (Day 266 and 267). Samples were taken from influent and effluent ports every hour for alkalinity, pH, temperature, dissolved oxygen, turbidity, iron, manganese and radon. Hourly measurements of total flow were also taken. Gamma readings, microbial numbers, uranium and radium analyses were not conducted during the diurnal studies.

As observed during Phase II, there was no change ($\alpha = 0.05$ and 0.10, StT) in pH and temperature as the water passed through the GAC units (Figures 50 and 51). The alkalinity (Figure 50) remained relatively constant during the diurnal study. The DO significantly ($\alpha = 0.05$ and 0.10, StT) decreased through the GAC system (Figure 51) by an amount comparable to that observed during Phase II. The turbidity was in the same low range observed during Phase II (Figure 52) and was significantly removed ($\alpha = 0.05$ and 0.10) through the units (0.09 ± 0.02 to 0.05 ± 0.01) on Day 266, but not Day 267. The change was relatively small and somewhat variable and the data were at or below the detection limit (0.05 NTU). The iron and manganese data were all below detection.

The water flowrate during this period was higher than the average (Figure 53). On Day 266, the flow averaged 49 ± 12 m^3/day (EBCT = 38.7 min) and was not significantly different ($\alpha = 0.05$ and 0.10, StT) than that observed on Day 267 (50 ± 8 m^3/day). There was some variation during the day, as would be expected in a small community. The influent radon activity was significantly different ($\alpha = 0.05$ and 0.01, StT) on the 2 days (Figure 53). On Day 266, it averaged 190,155 ± 3,739 pCi/L, while it was 165,013 ± 7,542 pCi/L on Day 267. In addition on Day 267, there was a steady decrease in influent activity over time. In spite of the differences in the influent activity, the effluent

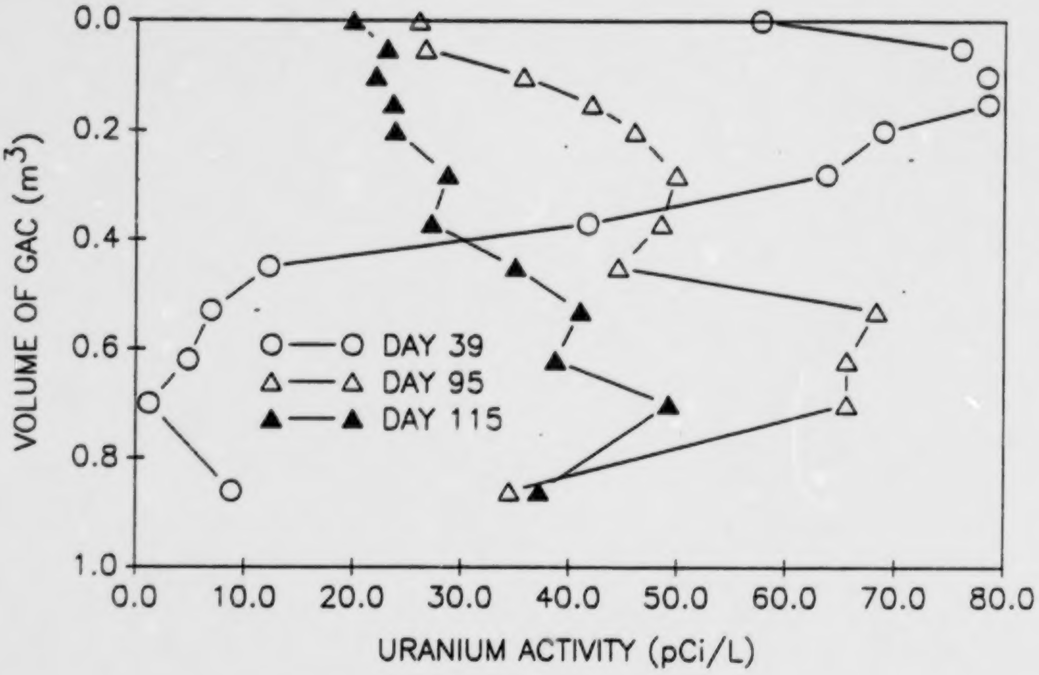


Figure 49. Phase II - Amherst, NH GAC system. Representative profiles of uranium through the GAC system.

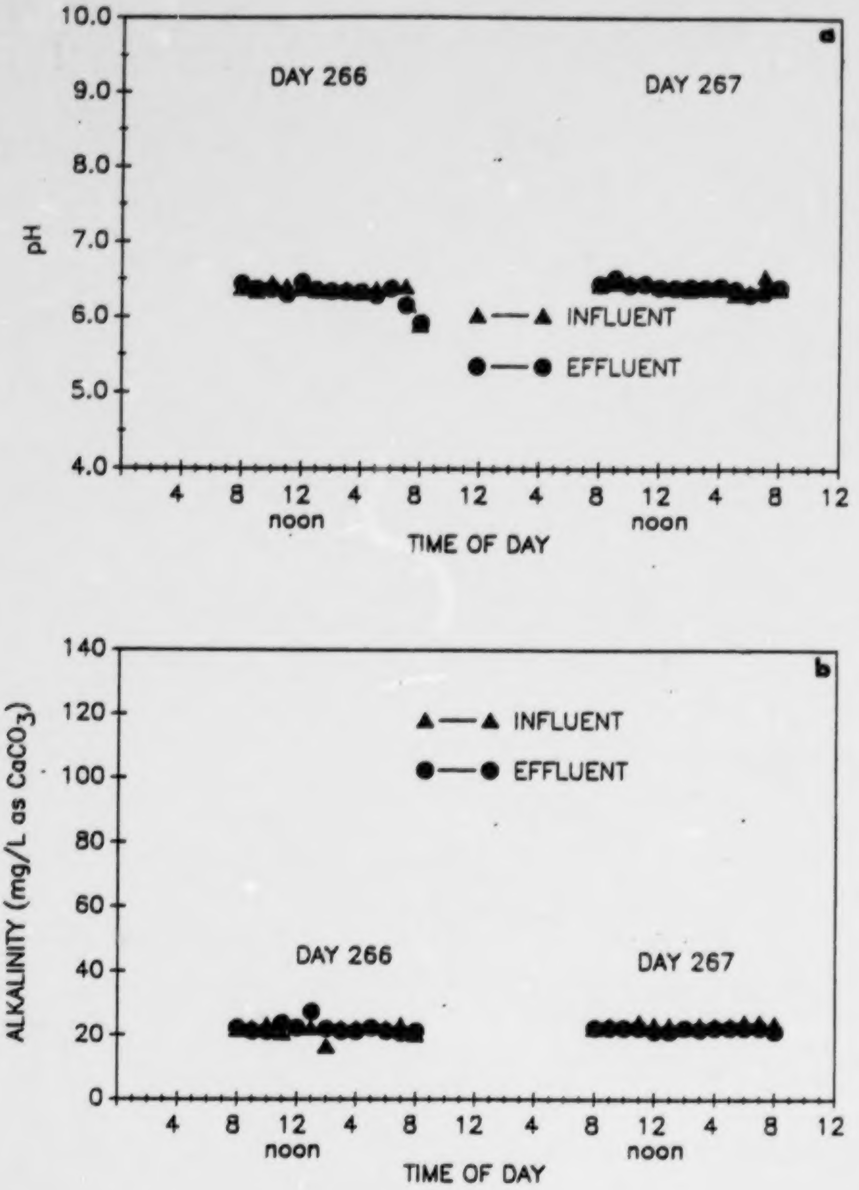


Figure 50. Diurnal variations - Mont Vernon, NH GAC system. pH (a) and alkalinity (b) data.

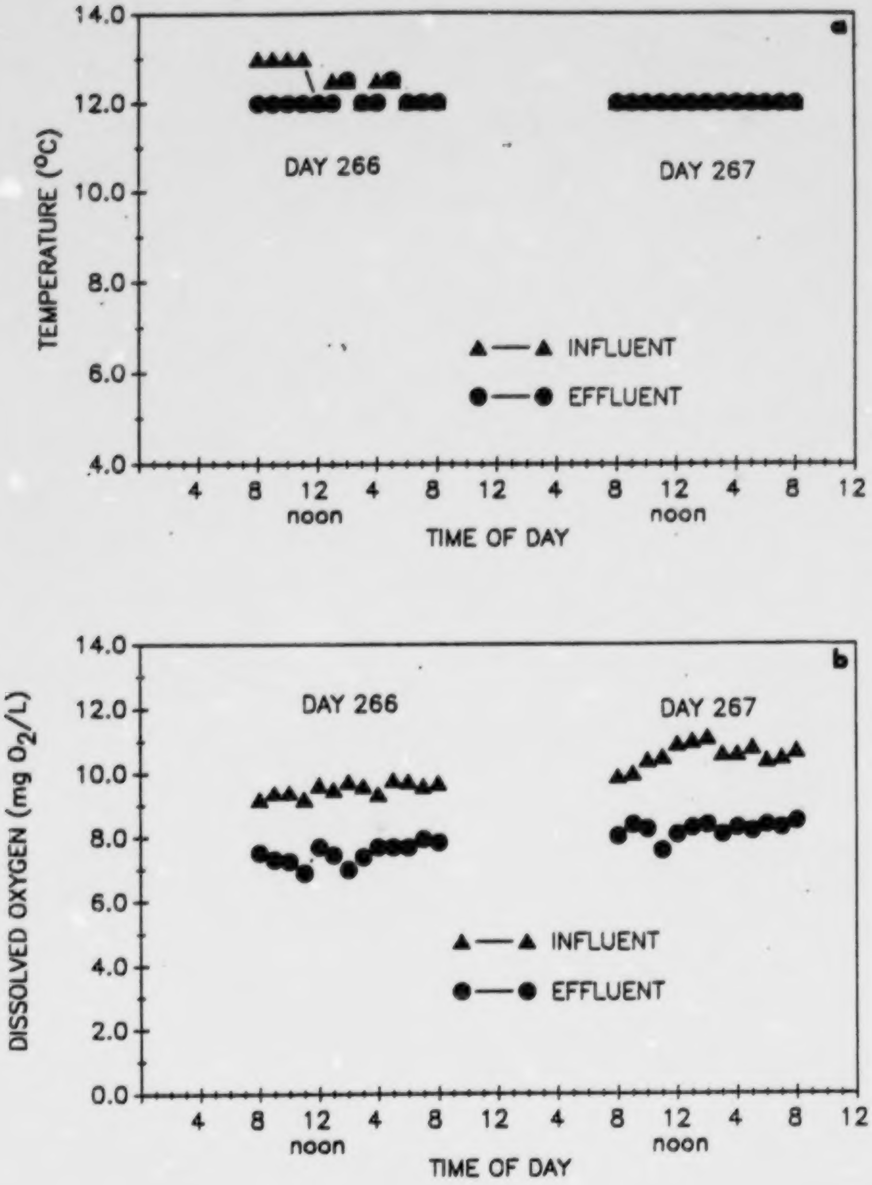


Figure 51. Diurnal variations - Mont Vernon, NH GAC system. Temperature (a) and dissolved oxygen (b) data.

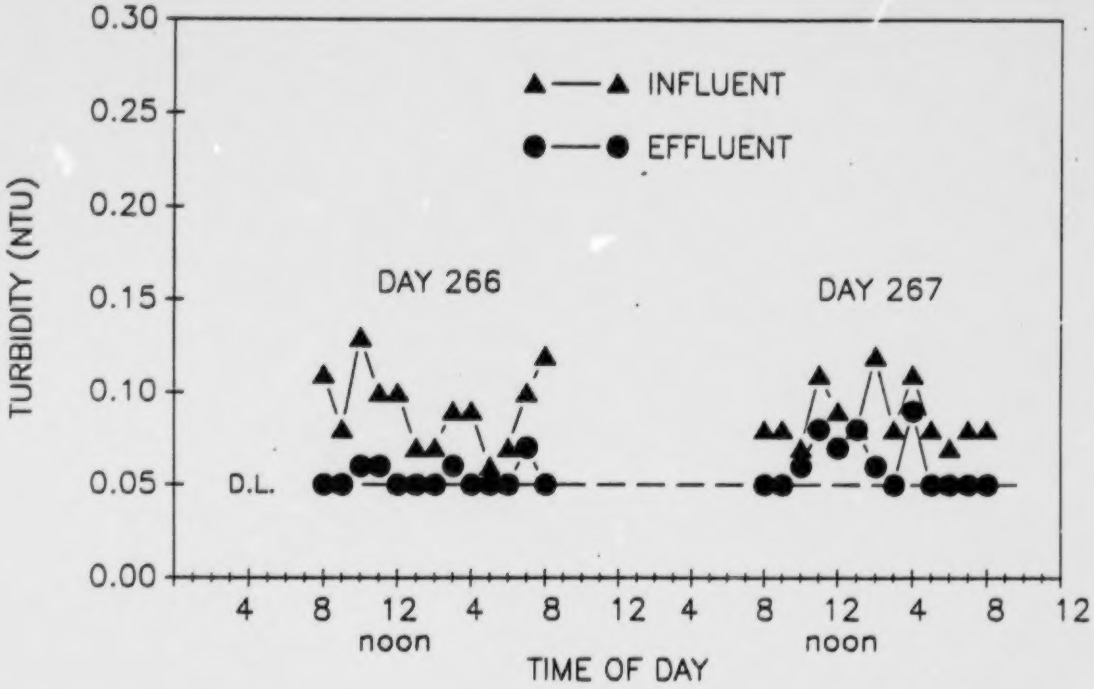


Figure 52. Diurnal variations - Mont Vernon, NH GAC system. Turbidity data (detection limit = 0.05 NTU).

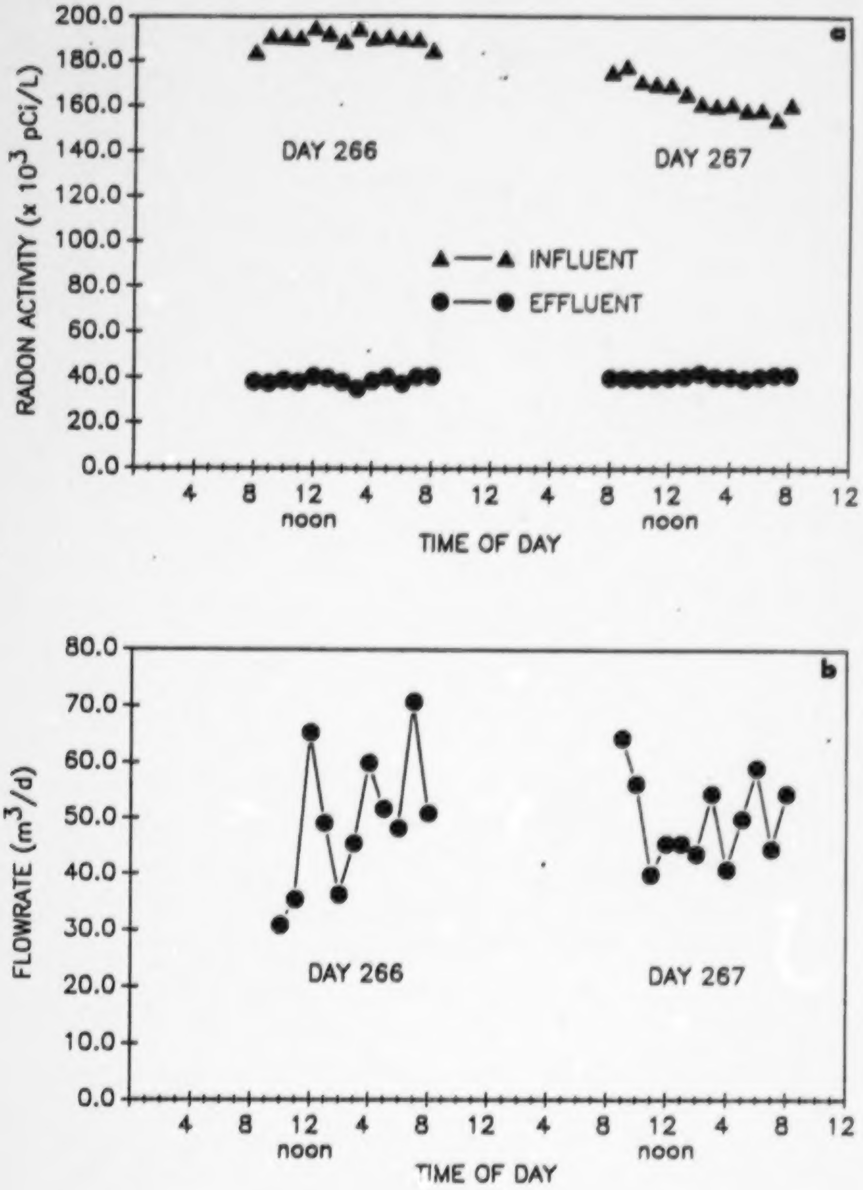


Figure 53. Diurnal variations - Mont Vernon GAC system. Radon activity (a) and water flowrate (b) data.

radon activity on the two days was not significantly different ($\alpha = 0.05$ and 0.10, StT). Although fluctuations in the influent conditions to the GAC systems were dampened during Phase I and II, the diurnal data indicate that the Mont Vernon units did not produce a better effluent when the influent radon activity decreased by ~25,000 pCi/L for a given water flowrate. The K_{ss} calculated for Day 266 ($2.5 \pm 0.6\ hr^{-1}$) was similar to the Phase II K_{ss} ($2.4 \pm 0.7\ hr^{-1}$). The Day 267 K_{ss} was, however, lower ($2.0 \pm 0.7\ hr^{-1}$). The fact that the effluent quality did not improve may have resulted because (i) the GAC system was not at steady state, (ii) radon was desorbing in response to the decrease in influent activity, and/or (iii) a water quality parameter(s) not monitored during the diurnal study was affecting radon adsorption. Further studies would need to be conducted to determine why the GAC system did not respond to the decreased influent loading.

High Flow--

The effect of high flow on the Mont Vernon system was investigated in July, 1987 (Day 281). Influent and effluent samples and measurements for flow, alkalinity, pH, temperature, turbidity, dissolved oxygen, microbial populations, iron, manganese and radon were taken every hour for 3 hr. The wells would not sustain a continuous flowrate at that level without depleting the community's water storage. As a result, the study was stopped after 3 hr. Radon samples were taken from all ports.

As observed in Phase II, there was no change ($\alpha = 0.05$ and 0.10, StT) in pH (Figure 54), alkalinity (Figure 54), and temperature (Figure 55) through the GAC units during the high flow study. DO was significantly reduced through the filter ($\alpha = 0.05$ and 0.10, StT) after 1 hr of operation, but not before that time (Figure 55). Turbidity remained low and was not significantly removed through the filter ($\alpha = 0.05$ and 0.01, StT). These changes are similar to those observed during Phase II. Iron and manganese were both below detection. The microbial numbers were variable (Figure 56) as observed in Phase II, but generally higher in the effluent than in the influent.

The flowrate during this part of the GAC evaluation averaged 73 ± 9.0 m^3/day (EBCT = 26.3 min) which was high compared to the flowrate during most of the study, but not during the period of June to August 1987. The flowrate for the high flow study was approximately 3 times the design of 25 m^3/day. Over the 3 hr period, the influent and effluent radon activity averaged $212{,}781 \pm 11{,}164$ pCi/L and $52{,}720 \pm 574$ pCi/L, respectively. These remained very consistent as did radon removal through the units (Figure 57). The K_{ss} for this period averaged $3.18 \pm 1.39\ hr^{-1}$ compared with the $2.4 \pm 0.72\ hr^{-1}$ value during Phase II. This is closer to the $3.02\ hr^{-1}$ at 6-10°C K_{ss} found previously with this GAC (Lowry and Lowry, 1987). Overall, the GAC system performed well during this study, producing a constant effluent radon activity in response to a high radon loading.

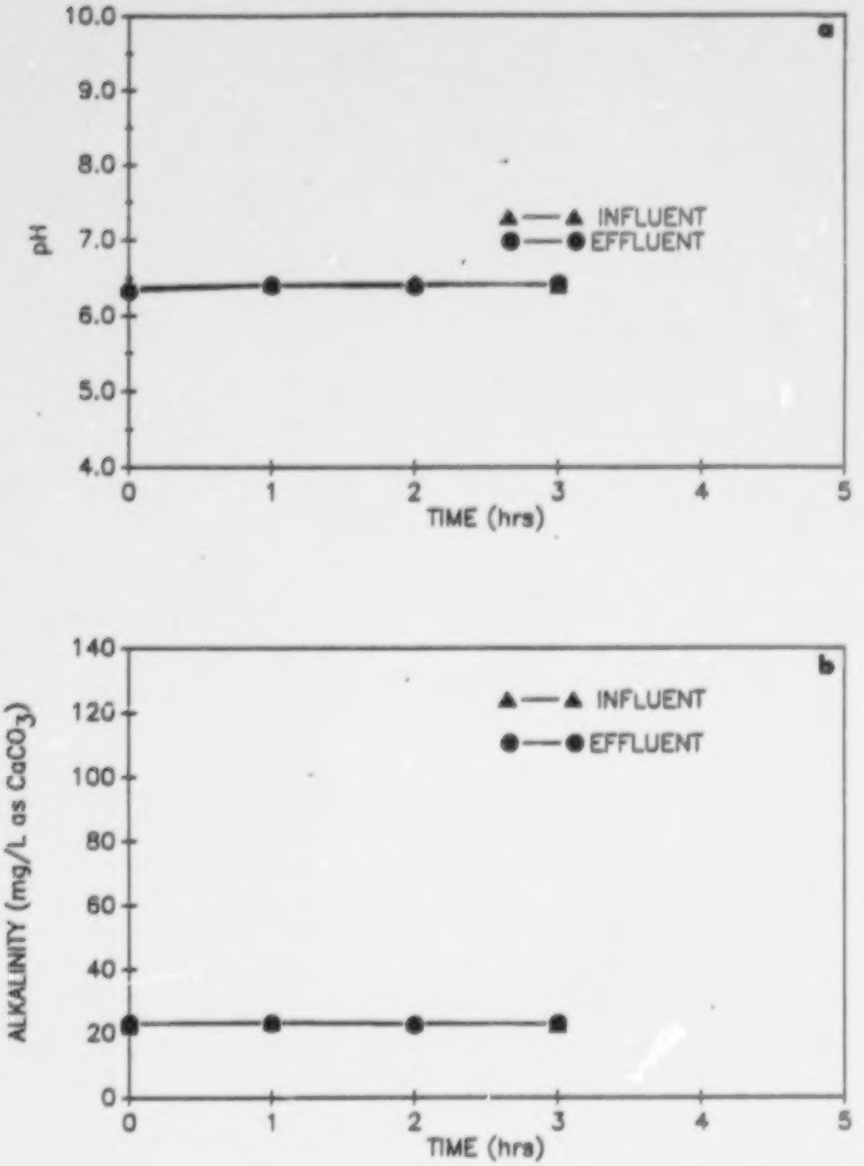


Figure 54. High flowrate - Mont Vernon, NH GAC system. pH (a) and alkalinity (b) data.

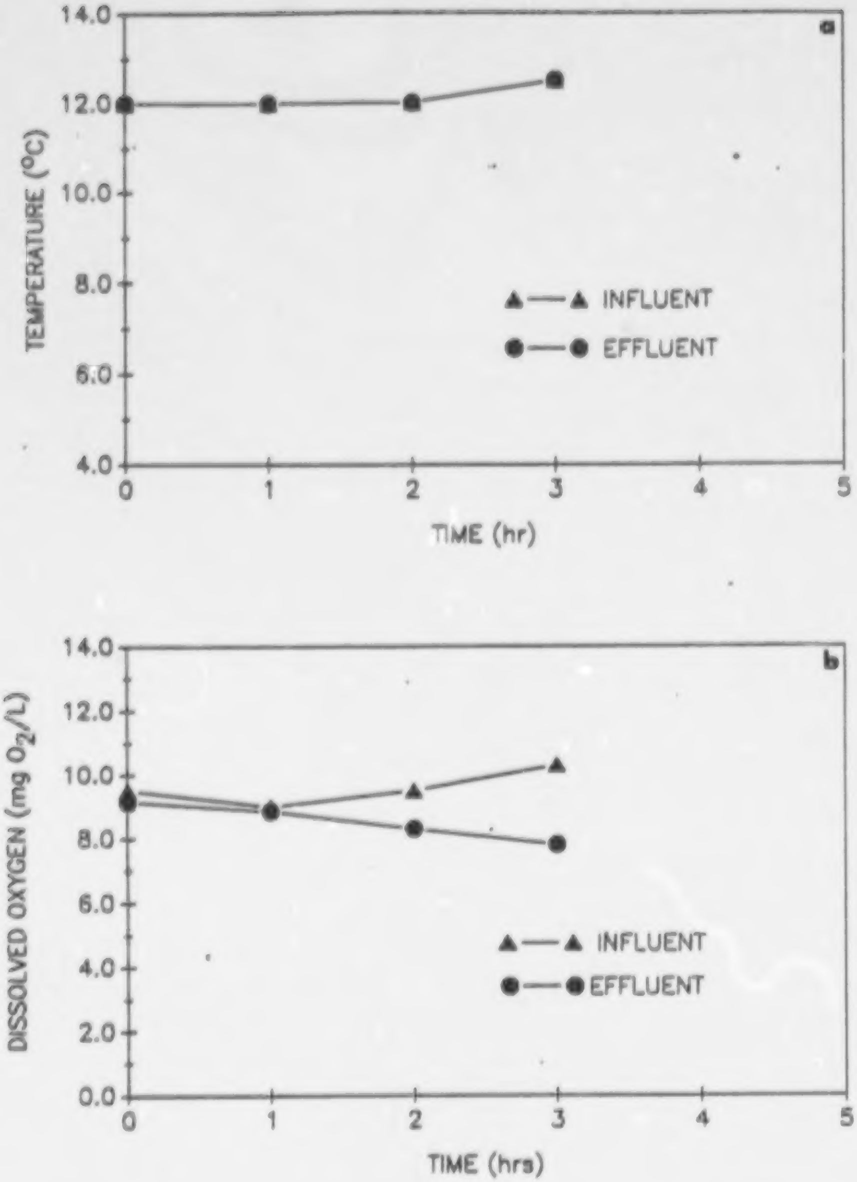


Figure 55. High flowrate - Mont Vernon, NH GAC system. Temperature (a) and dissolved oxygen (b) data.

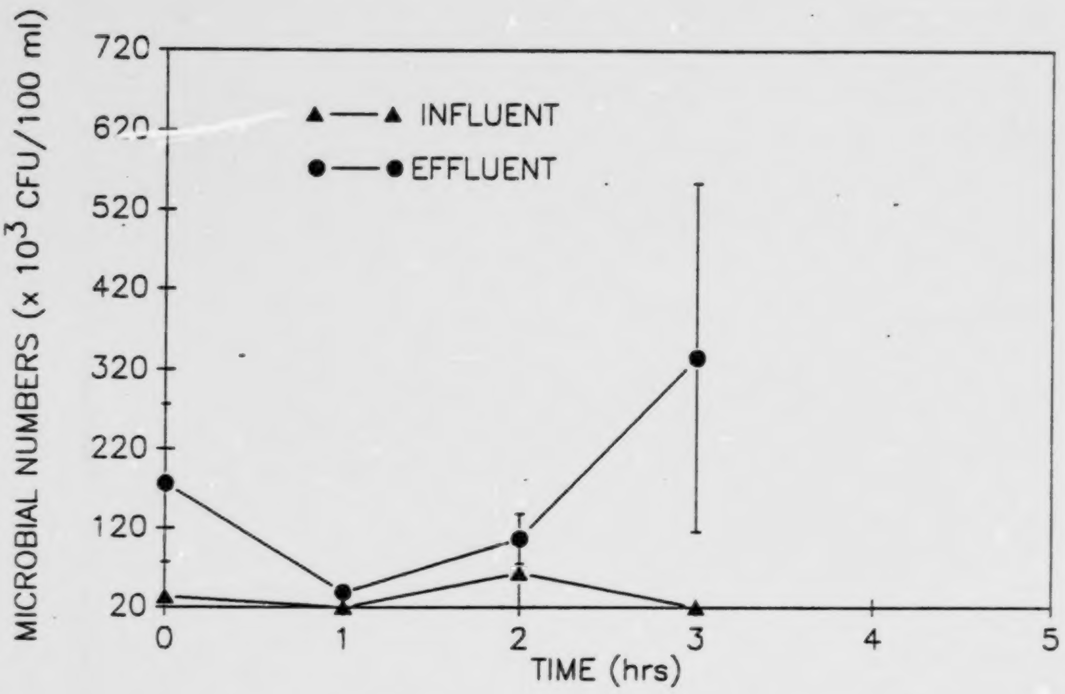


Figure 56. High flowrate - Mont Vernon, NH GAC system. Microbial numbers (detection limit = 20,000 CFU/100 mL).

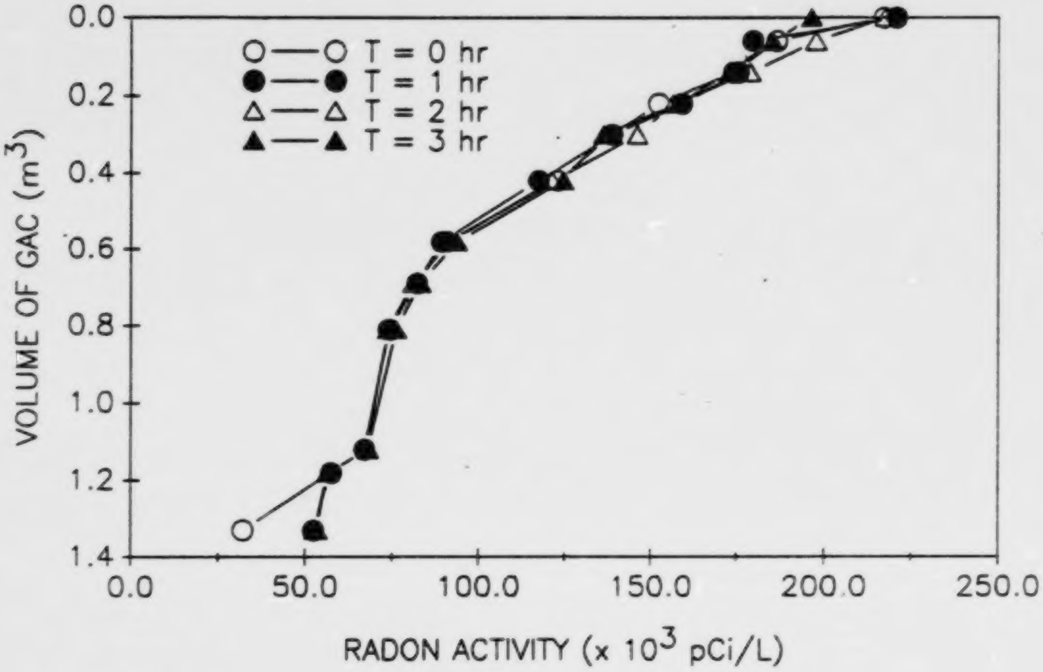


Figure 57. High flovrate - Mont Vernon, NH GAC system. Radon activities through the GAC system.

Coring--

Cores of the GAC material were obtained from the Mont Vernon and Amherst systems in late July 1987. The Mont Vernon system had been operating for 293 days before it was cored. The Amherst GAC unit had only operated for 122 days and it had been out of service for 171 days prior to coring. Each core was sectioned, and composite samples of each section were collected for percent moisture content, microbial populations, iron, manganese, uranium radionuclides, radium-226 and lead-210 analyses. Uranium and radium analyses were done by the State of New Hampshire and the University, while only the University made lead-210 measurements. All data were calculated in kg per dry weight of GAC (based on percent moisture content) and reflect a composite of the GAC volumes shown in Table 6. Samples of virgin GAC were also analyzed and the values obtained were subtracted out of all data presented.

Mont Vernon site--Three cores of GAC #1 and two cores of GAC #2 were taken from the Mont Vernon system after 293 days of operation. Manganese was below detection except in the bottom 0.27 m^3 of GAC in GAC #1 (2.7 mg/kg) and the bottom 0.41 m^3 of GAC #2 (2.9 mg/kg) (Figure 58). The highest iron accumulation was found in the top of GAC #2 (Figure 58). The iron accumulation pattern is likely the result of dissolved (ferrous) iron entering the filter, undergoing oxidation as it passed through the upper part of GAC #1 and being deposited as an insoluble (ferric) iron complex (e.g., $Fe(OH)_3$) in the bottom of GAC #1 and the top of GAC #2. It was impossible to compare the coring data to a theoretical removal as the iron concentrations in the water were usually below detection. However, the data indicate that accumulation can occur even with small changes in iron concentration because of the large volume of water passing through the units over time. As a result, backwashing may be required to prevent significant headloss. Microorganisms were growing throughout the two units (Figure 59), but were most concentrated in the bottom of GAC #2. The microbial numbers found in the GAC units at Mont Vernon are comparable to those reported by Wilcox et al. (1983), who found that the CFU per gram of GAC ranged from 3×10^5 to 6×10^7.

The estimated uranium adsorbed corresponded fairly well to the coring data (Table 7) considering these estimates were based on an average uranium activity and total flow through the unit and did not take into account the actual variations occurring at the site. It was not possible to do an accurate mass balance on any of the elements sampled because of the natural variation in influent water quality and flowrate occurring in the full-scale small community facilities.

The uranium 238 and 235 profiles (Figure 60) and the uranium removal profile obtained during Phase II sampling (Figure 47) both indicate uranium was removed exponentially through the GAC system. There was a progressive movement of the uranium adsorption front through the filter observed in Phase II (Figure 47) and the coring data indicated that the GAC was approaching exhaustion with respect to uranium so that eventually no additional removal would be occurring. The data indicate that uranium was being retained by the GAC and not desorbed, however, studies with careful regulation of influent

TABLE 6. GAC VOLUMES COMPOSITED IN CORING EVENT

Site	Volumes Composited (m^3)
Mont Vernon	0.00 - 0.14*
	0.14 - 0.30*
	0.30 - 0.57*
	0.57 - 0.69+
	0.69 - 0.93+
	0.93 - 1.33+
Amherst	0.00 - 0.20
	0.20 - 0.45
	0.45 - 0.85

* GAC #1.

+ GAC #2.

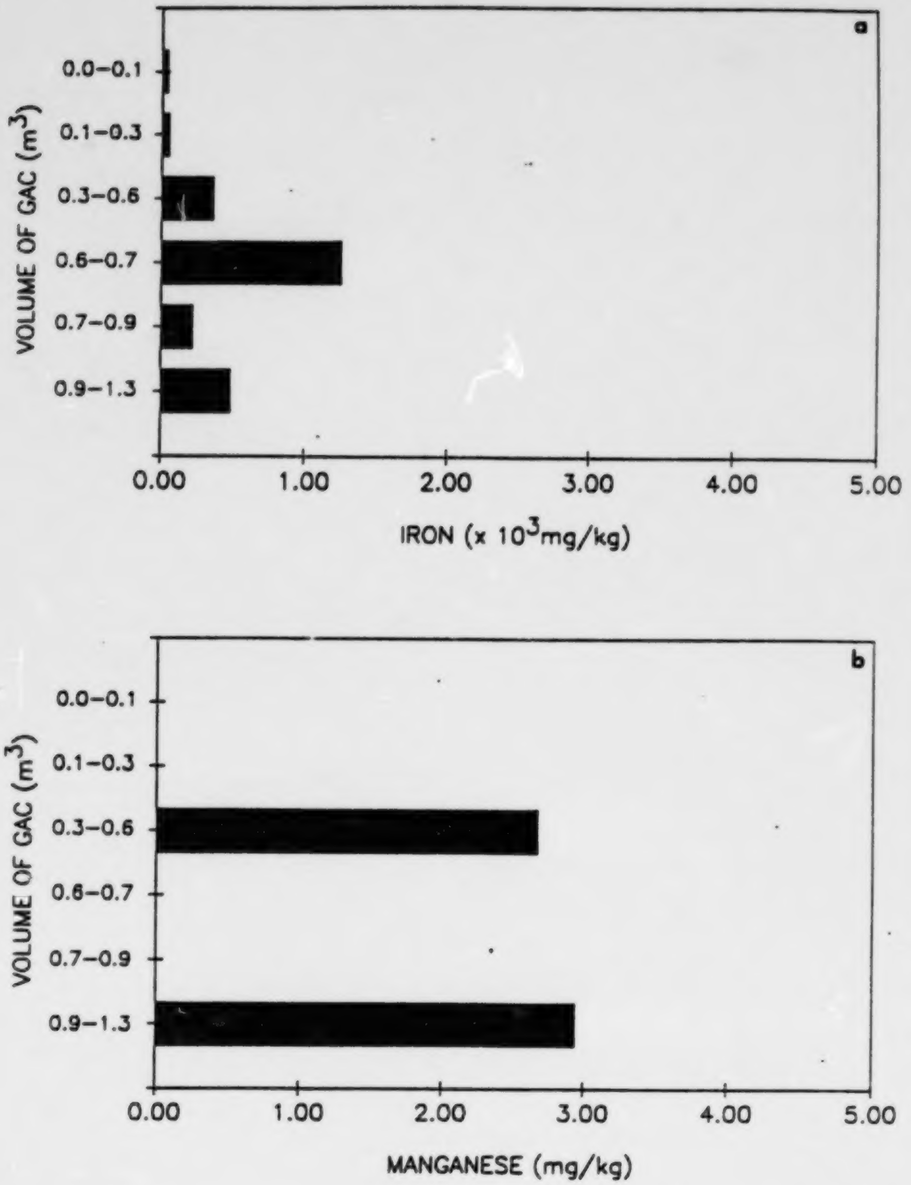


Figure 58. Coring event - Mont Vernon, NH GAC system. Iron (a) and manganese (b) found in GAC system.

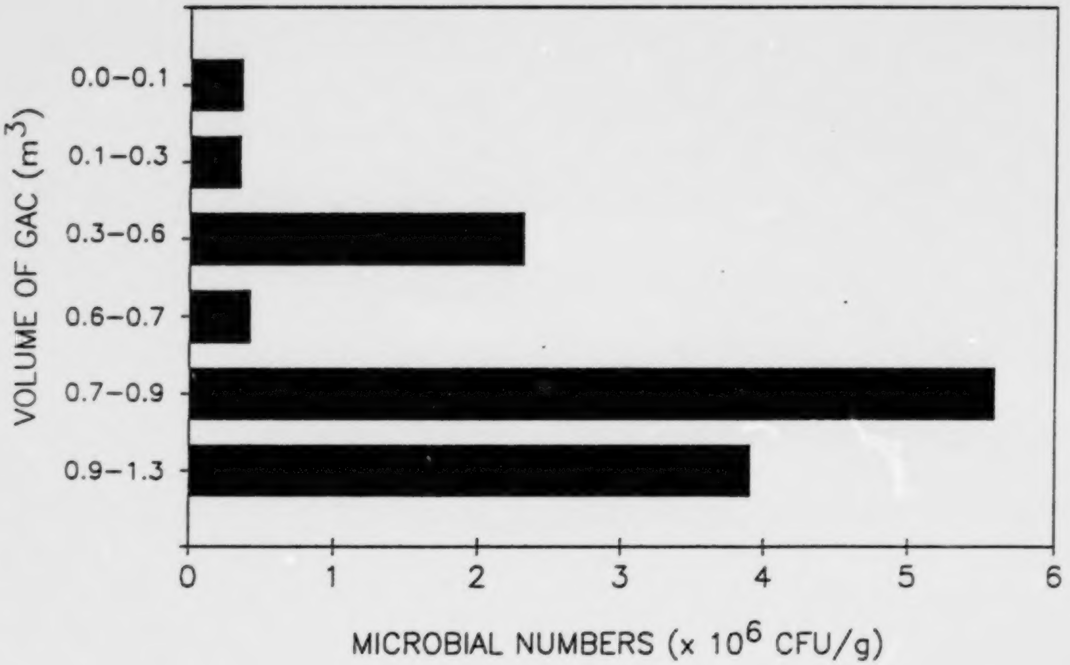


Figure 59. Coring event - Mont Vernon, NH GAC system. Microbial numbers found in the GAC system.

TABLE 7. COMPARISON OF THEORETICAL URANIUM[*] ADSORBED TO MONT VERNON CORING DATA

Volume of GAC (m^3)	Theoretical Adsorbed[+] (pCi/kg)[**]	Coring Results (pCi/kg)	
		State[++]	University[***]
0.0 - 0.14	1,542,719	905,200	549,200
0.14 - 0.30	1,038,363	457,100	313,000
0.30 - 0.57	732,371	425,200	264,800
0.57 - 0.69	123,559	92,100	85,560
0.69 - 0.93	17,074	17,500	5,000
0.93 - 1.33	~0	598	290

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[*] Coring data includes uranium-238 and uranium-235. Theoretical calculation based on total uranium as assessed by EPA Method 908.0 (EPA, 1980).

[+] Calculated using average total uranium activity and water flowrate measured during operating period.

[**] pCi/kg dry weight of GAC, based on percent moisture data.

[++] State of New Hampshire Division of Public Health Services Laboratory.

[***] University of New Hampshire Institute of Earth, Oceans and Space Laboratory.

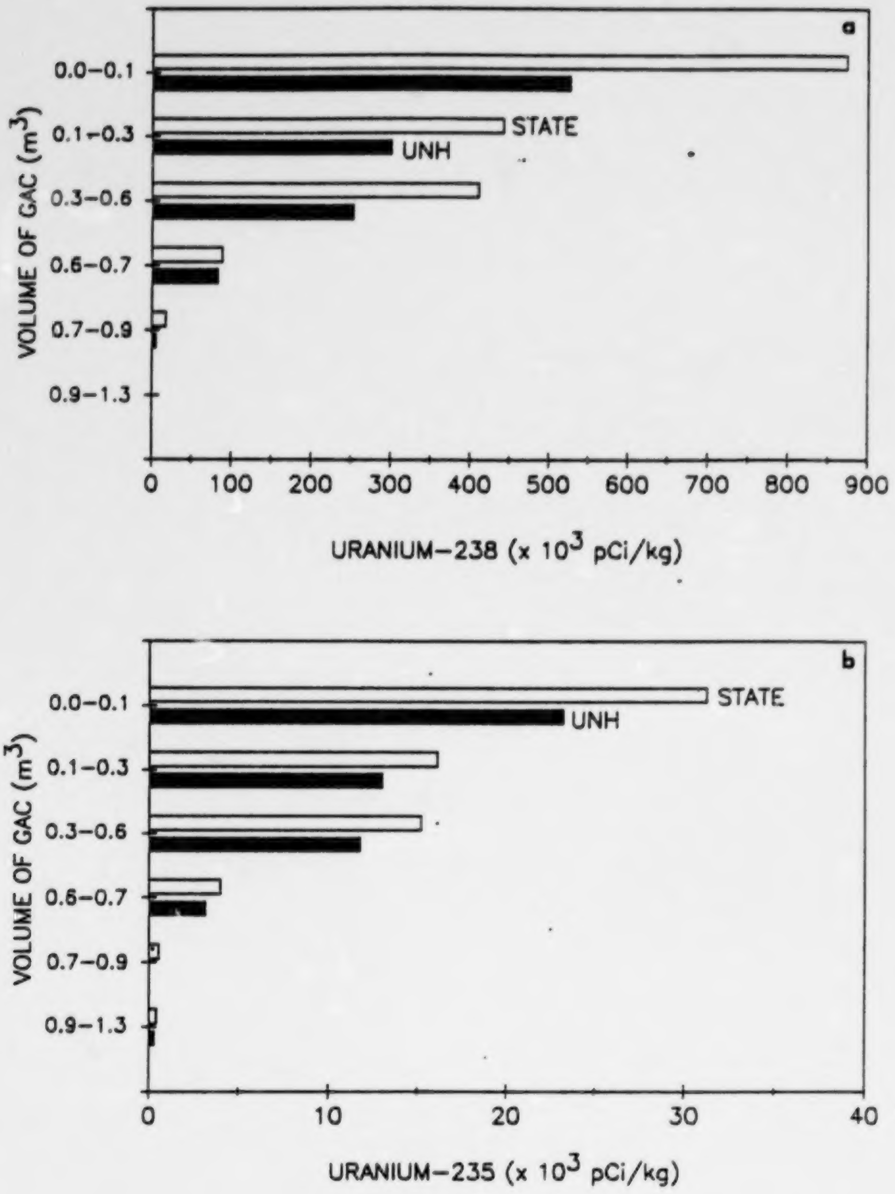

Figure 60. Coring event - Mont Vernon, NH GAC system. Uranium-238 (a) and uranium-235 (b) found in the GAC system.

activity, flowrate and water quality would be needed to verify this observation.

There was very little radium-226 data collected during Phase II, hence comparisons of theoretical adsorbed radium and the coring data could not be made. The cores (Figure 61) indicated that significant quantities of radium were found throughout the entire GAC system. Since the removal of radium by GAC adsorption alone is considered unfavorable, (Sorg and Logsdon, 1978) it is hypothesized that the radium found in the GAC cores may have been removed by adsorption or ion exchange reactions occurring with other solid phases (e.g., $Fe(OH)_3$, $Mg\ CO_3$) or organic matter deposited on the GAC.

The lead-210 coring data was indicative of the retention of radon progeny on the GAC. As observed for the radon removal profiles during Phase II (Figures 34 and 35), the sorbed lead-210 content decreased exponentially through the unit (Figure 61). A comparison of the theoretical lead-210 adsorbed and lead-210 coring data (Table 8) indicated that most, if not all of the radon progeny were retained by the GAC. As with the other comparisons, it was impossible to make a completely accurate mass balance because of variations in the raw water quality and flowrate. Further study under more controlled conditions would be required to demonstrate whether retention is complete or affected by variations in the raw water loading.

Rubin and Mercer (1981) reported that GAC adsorption capacities for (non-radioactive) lead ranged from 6.2×10^4 to 1.9×10^6 µg/kg at pH 6.5 and from 2.1×10^6 to 1.8×10^8 µg/kg at pH 8.0 based on Langmuir adsorption isotherms. Comparison of these data to the lead-210 coring results (Table 8) indicates that the GAC's capacity for lead-210 at Mont Vernon was at least 10^6 times higher than the actual lead-210 loading. This supports the hypothesis that lead-210 was completely retained by the GAC units. However, it should be noted that lead removal by GAC is highly dependent upon pH (Netzer and Hughes, 1984). Therefore, the ability of the GAC to retain lead-210 may be highly dependent on pH and raw water quality and should not be assumed without verification.

The GAC in both units, except for the bottom 0.41 m^3 of GAC #2, exceeds the limit for uranium-238 (58,410 pCi/kg; 2.5×10^{-5} Ci/m^3) as outlined by the New Hampshire Radiological Health Program de minimus regulations (1983). All materials which exceed de minimus levels are considered to be low level radioactive wastes. Ultimately, it appears all of the GAC at Mont Vernon would have exceeded the de minimus standards if monitoring had continued. All of the GAC is below the de minimus level (58,410 pCi/kg; 2.5×10^{-5} Ci/m^3) for uranium-235. All of the material presently exceeds the de minimus limit (44.39 pCi/kg; 1.9×10^{-8} Ci/m^3) for radium-226. The State of New Hampshire does not have a de minimus standard for lead-210. When the GAC at Mont Vernon needs to be replaced, it will have to be handled as a low level radioactive waste. This impacts the cost of replacement of the GAC system when it occurs and also has implications with respect to handling and public policy. It

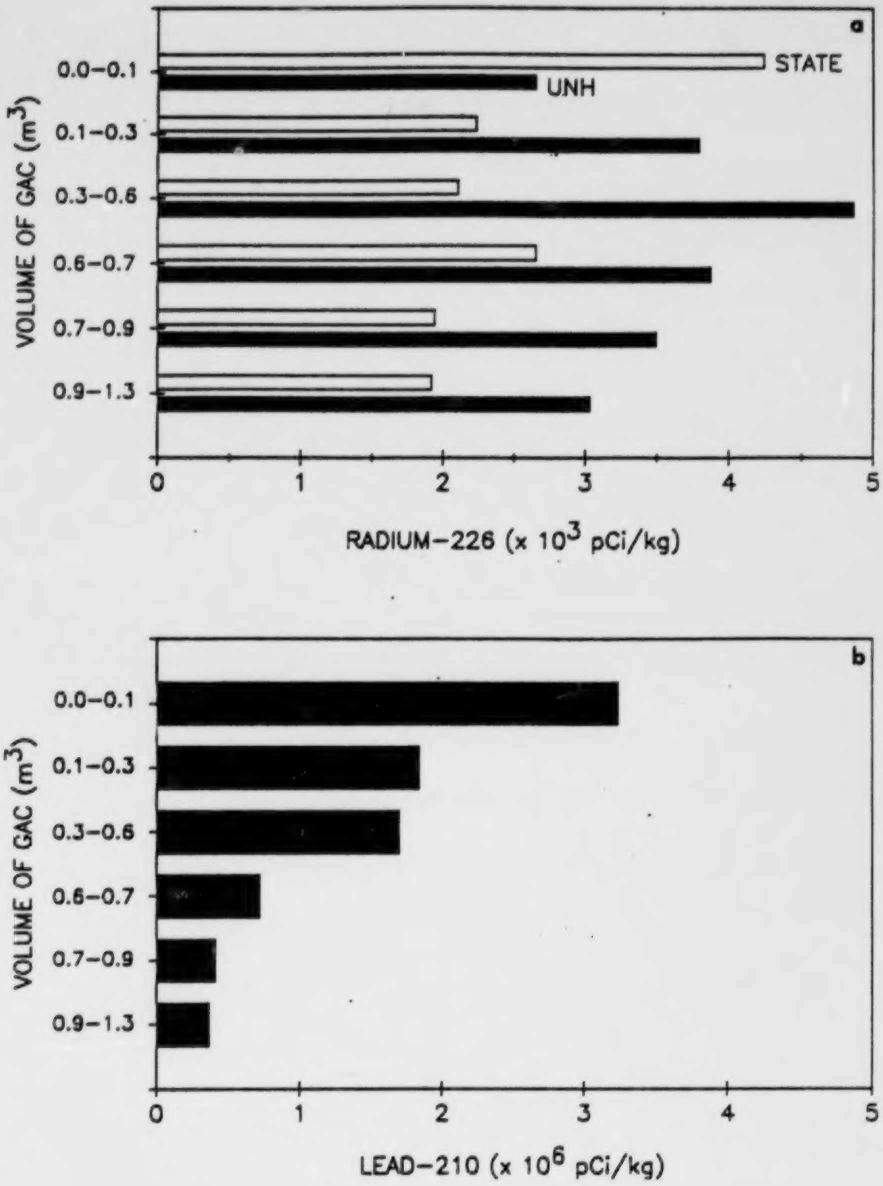

Figure 61. Coring event - Mont Vernon, NH GAC system. Radium-226 (a) and lead-210 (b) found in the GAC system.

TABLE 8. COMPARISON OF THEORETICAL LEAD-210[*] ADSORBED TO MONT VERNON LEAD-210 CORING DATA

Volume of GAC (m^3)	Total Radon-222 Removed (pCi/kg)[+]	Lead-210 Theoretical Adsorbed (pCi/kg)[+]	Lead-210 Coring Results (pCi/kg)[+]	Lead-210 Coring Results (µg/kg)
0.0 - 0.14	1.12×10^{10}	5.57×10^6	3.43×10^6	0.045
0.14 - 0.30	6.54×10^9	3.26×10^6	1.94×10^6	0.026
0.30 - 0.57	4.63×10^9	2.31×10^6	1.80×10^6	0.024
0.57 - 0.69	2.59×10^9	1.29×10^6	7.57×10^5	0.010
0.69 - 0.93	2.89×10^9	1.44×10^6	4.28×10^5	0.006
0.93 - 1.33	1.35×10^9	6.75×10^5	3.83×10^5	0.005

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[*] See Appendix C for methods of calculation. The total theoretical lead-210 activity in GAC #1 = 8.24×10^8 pCi and GAC #2 = 3.32×10^8 pCi. The total measured lead-210 in GAC #1 = 5.46×10^8 pCi and GAC #2 = 1.50×10^8 pCi (based on coring results).

[+] pCi/kg dry weight of GAC, based on percent moisture data.

should be noted that the State of New Hampshire has stringent de minimus standards and that in many other states this material would not be considered to be a low level radioactive waste.

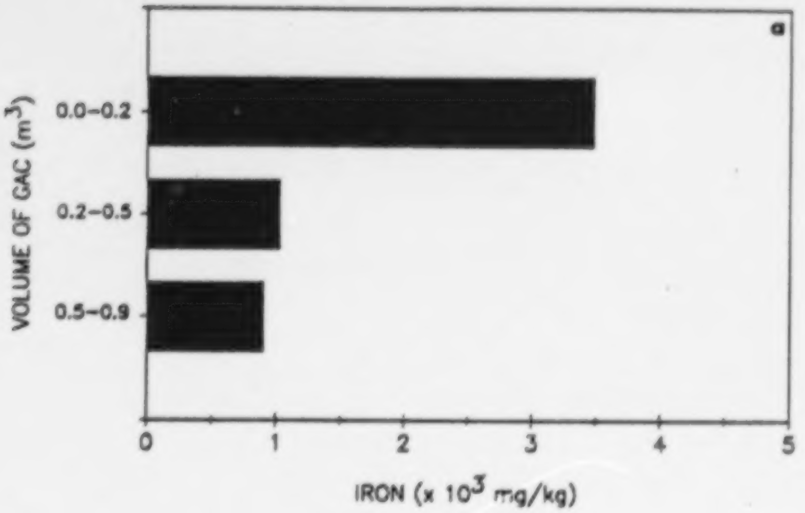
Amherst site--Two cores were taken of the Amherst GAC material. The microbial populations were not monitored because the unit had been out of service for approximately 6 months when the coring event occurred. Substantial amounts of manganese and iron (Figure 62) had accumulated. The data showed higher accumulations of these metals at the top of the filter with a decreasing trend through the unit, probably because these metals were entering the filter as precipitates due to oxidation in the atmospheric storage tank prior to the GAC. The accumulation of iron and manganese was greater than the accumulation at Mont Vernon considering the Amherst unit had operated 6 months less than the Mont Vernon GAC. This was expected as the raw water concentrations of iron and manganese and the flowrate were higher at Amherst.

The coring data for uranium 238 and 235 (Figure 63) and the Phase II uranium data (Figure 49) at Amherst both indicated that the GAC did not effectively adsorb uranium. In comparison to the coring data obtained at Mont Vernon, the Amherst GAC adsorbed very little uranium. The discrepancy may be explained by the different pH's at the two sites (Amherst = 8.03 ± 0.14; Mont Vernon = 6.5 ± 0.2). As previously discussed, uranium species are predominately anionic soluble carbonate complexes in natural waters from pH 7 to 10 (Sorg, 1988), while below pH ~ 6.8, the neutral UO_2CO_3 species predominates. Therefore, it is assumed that the poorly adsorbed anionic species predominated at Amherst whereas the favorably adsorbed neutral species predominated at Mont Vernon, which would account for the observed differences.

The radium-226 coring data (Figure 64) indicate that this radionuclide was retained by the GAC in a manner similar to that at Mont Vernon. A comparison with the theoretical radium-226 adsorbed could not be made because only one analysis was made during Phase II at Amherst.

The lead-210 coring data (Figure 64) again showed an exponential pattern similar to the radon removal observed during Phase II (Figure 37). The theoretical comparison (Table 9) yielded similar values considering variation in radon activity and flowrate were not known. The amount of lead-210 adsorbed was much less than that observed at Mont Vernon because the Amherst GAC unit was only operated for 4 months.

The Amherst GAC exceeded the State of New Hampshire de minimus levels for uranium-238 (58,410 pCi/kg; 2.5×10^{-5} Ci/m³) and radium-226 (44.39 pCi/kg; 1.9×10^{-8} Ci/m³) throughout the filter. As a result, all of the GAC at Amherst was classified as a low level radioactive waste. The uranium-235 concentrations were below de minimus standards (58,410 pCi/kg; 2.5×10^{-5} Ci/m³) and lead-210 is not regulated in New Hampshire.



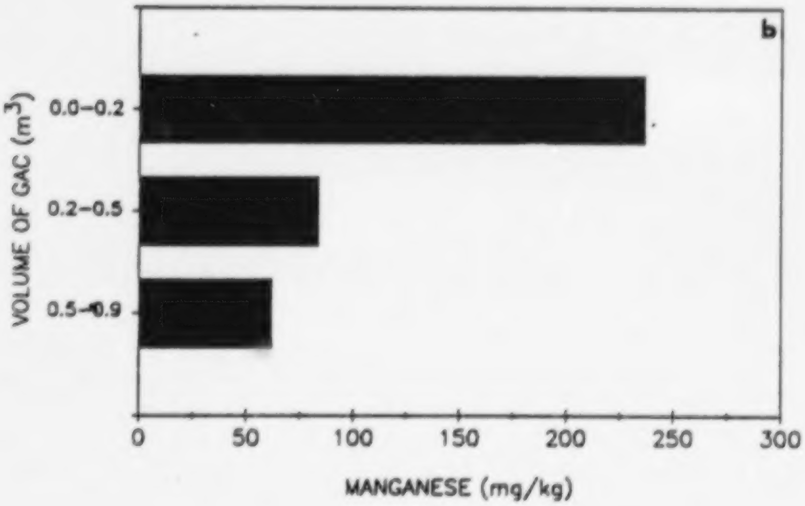
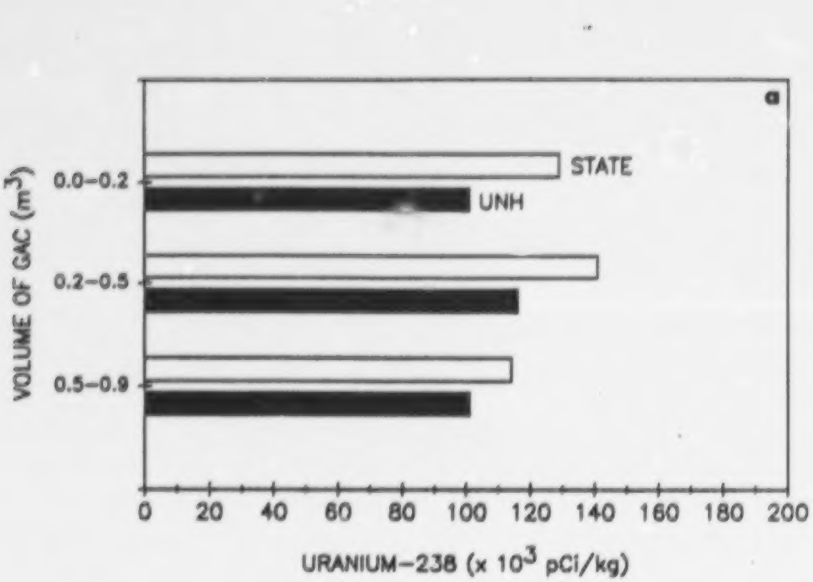


Figure 62. Coring event - Amherst, NH GAC system. Iron (a) and manganese (b) found in the GAC system.



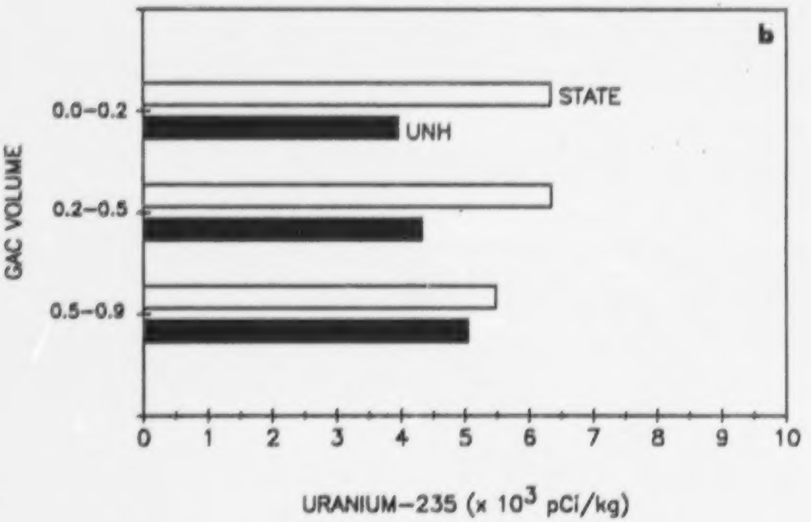
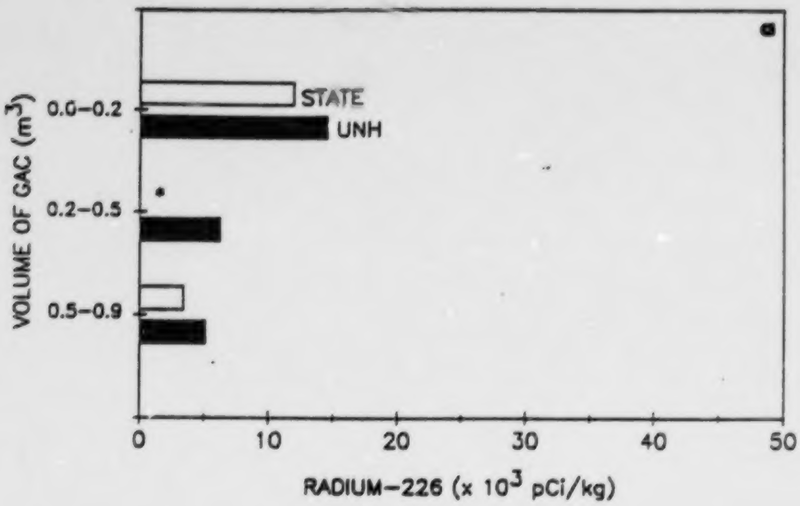


Figure 63. Coring event - Amherst, NH GAC system. Uranium-238 (a) and uranium-235 (b) found in the GAC system.



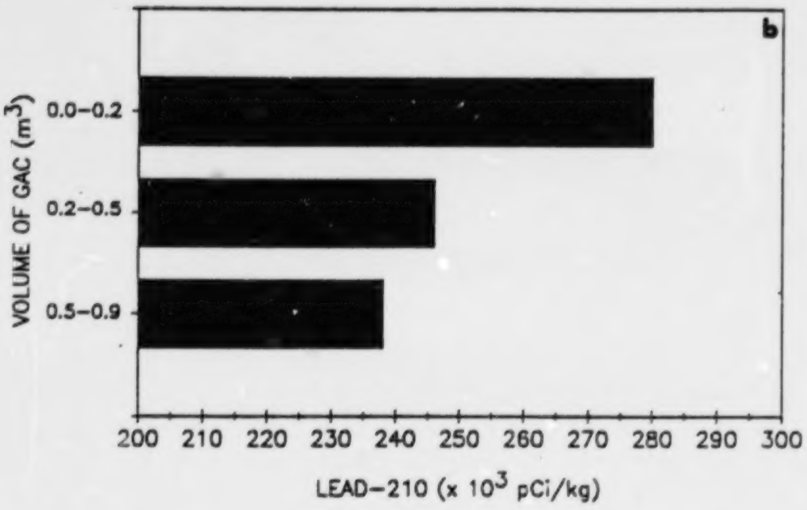


Figure 64. Coring event - Amherst, NH GAC system. Radium-226 (a) and lead-210 (b) found in the GAC system. (* Data from State laboratory unavailable.)

TABLE 9. COMPARISON OF THEORETICAL LEAD-210* ADSORBED TO AMHERST LEAD-210 CORING DATA

Volume of GAC (m^3)	Total Radon-222 Removed (pCi/kg)+	Lead-210 Theoretical Adsorbed (pCi/kg)+	Lead-210 Coring Results (pCi/kg)+	Lead-210 Coring Results (µg/kg)
0 - 0.20	1.48×10^9	7.39×10^5	2.97×10^5	0.004
0.20 - 0.45	8.66×10^8	4.32×10^5	2.59×10^5	0.003
0.45 - 0.85	4.93×10^8	2.46×10^5	2.51×10^5	0.003

* See Appendix C for methods of calculation. The total theoretical lead-210 activity in GAC bed = 1.52×10^8 pCi. The total measured lead-210 in the GAC bed = 9.61×10^7 pCi (based on coring results).

+ pCi/kg dry weight of GAC, based on percent moisture data.

Backwashing--

The GAC filters at Mont Vernon were backwashed individually in January 1988 (Day 477) using the raw water. The original backwash rate proposed (12.2 m^3/m^2-hr) caused the GAC to start flowing out of the filter. The backwash rate was immediately decreased to approximately 55 m^3/day (5 m^3/m^2-hr for GAC #1 and 3.5 m^3/m^2-hr for GAC #2) which eliminated this problem. Each unit was backwashed for 10 min as originally proposed. Normally, backwashing flowrates for GAC range from 29-49 m^3/m^2-hr for 10 to 15 min (Montgomery, 1985), but these flows would have been too high to prevent loss of GAC from the Mont Vernon filter since it was filled near capacity (36-53 cm freeboard). Samples for pH, alkalinity, DO, turbidity, iron, manganese and microbial numbers were taken immediately before and 1.3 and 24 hr after backwashing (~1 and 18 empty bed volumes, respectively). Samples for radon activity were taken before and at 1.3, 2.6, 3.9, and 24 hr after backwashing (~1, 2, 3 and 18 empty bed volumes, respectively). Samples were also taken of the backwash water and those were analyzed for iron, manganese, uranium and radon.

The water exiting the filters during backwashing was initially very turbid, especially from GAC #1. After 10 min of backwashing, the water from GAC #2 was clear, but the GAC #1 water remained somewhat turbid. The backwash water from the first minute of operation contained 0.68 mg Fe/L and 0.14 mg Mn/L from GAC #2 and 129.5 mg Fe/L and 1.33 mg Mn/L from GAC #1. After 10 min of backwashing, the iron concentration in the backwash water from GAC #2 remained the same and the manganese was below detention. At the end of backwashing, the iron and manganese concentrations in the GAC #1 backwash water were 4.37 mg/L and 0.11 mg/L, respectively. The backwashing had a major impact on removing iron and manganese precipitates from GAC #1, where they appeared to be most concentrated. These data seem to contradict those obtained from the coring (Figure 58) which indicated most of the iron was in the top of GAC #2. It is possible that the new well at Mont Vernon, installed after coring and in operation 3.5 months before backwashing contained higher amounts of iron which precipitated in the top of GAC #1. Another plausible explanation for the results might be the difference between iron associated with the pore water (i.e., removed by backwashing) and that which is adsorbed to the GAC (i.e., measured by coring). The uranium (~500 pCi/L) and radon (~28,000 - 43,000 pCi/L) activities measured in the backwash water were similar to those in the raw water from the new well at Mont Vernon indicating that these radionuclides were not desorbing from the GAC.

There was no significant change in the pH (Figure 65), alkalinity (Figure 65) and dissolved oxygen (Figure 66) obtained before and after backwashing (α = 0.05 and 0.10, StT). The turbidity profile improved primarily in the top 0.22 m^3 of GAC #1 after 24 hr with a decrease in turbidity from approximately 5.0 NTU to 0.10 NTU, showing a positive effect of backwashing. The impact of backwashing on the iron and manganese profiles (Figure 67) was not as positive. The backwashing appeared to move iron and manganese precipitates to the top of GAC #1, but did not improve the concentrations of these metals over 24 hr. There was some change in the microbial numbers (Figure 68) in the top of the GAC #1 after backwashing, however, caution must be used when interpreting these data because of the variability in microbial counts normally obtained from GAC filters.

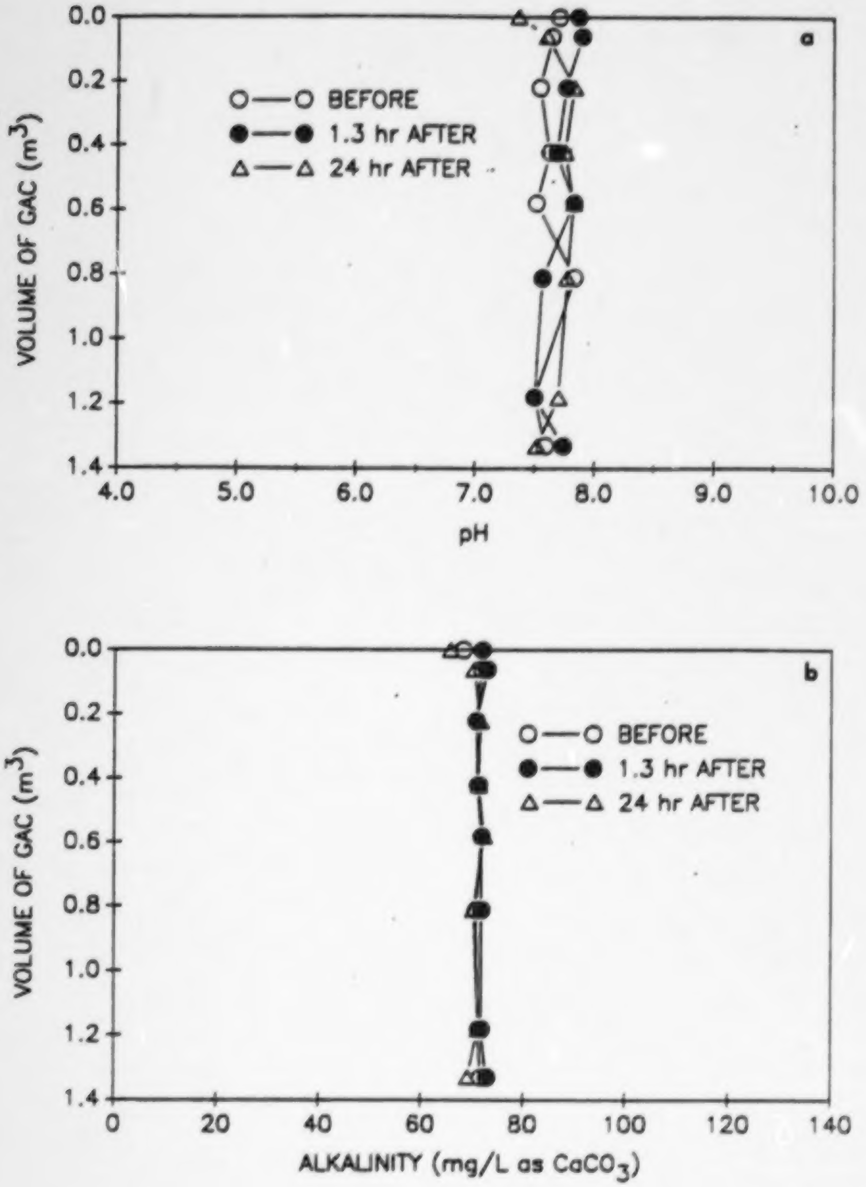


Figure 65. Backwash event - Mont Vernon, NH GAC system. pH (a) and alkalinity (b) profiles before and after backwashing.

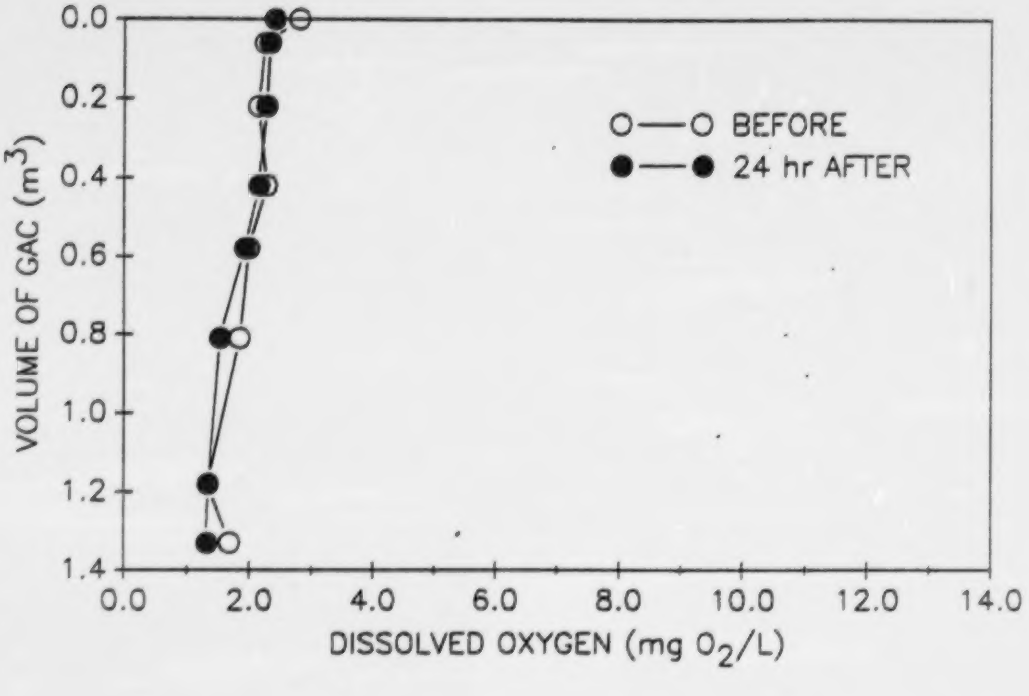


Figure 66. **Backwash event - Mont Vernon, NH GAC system. Dissolved oxygen profile before and after backwashing.**

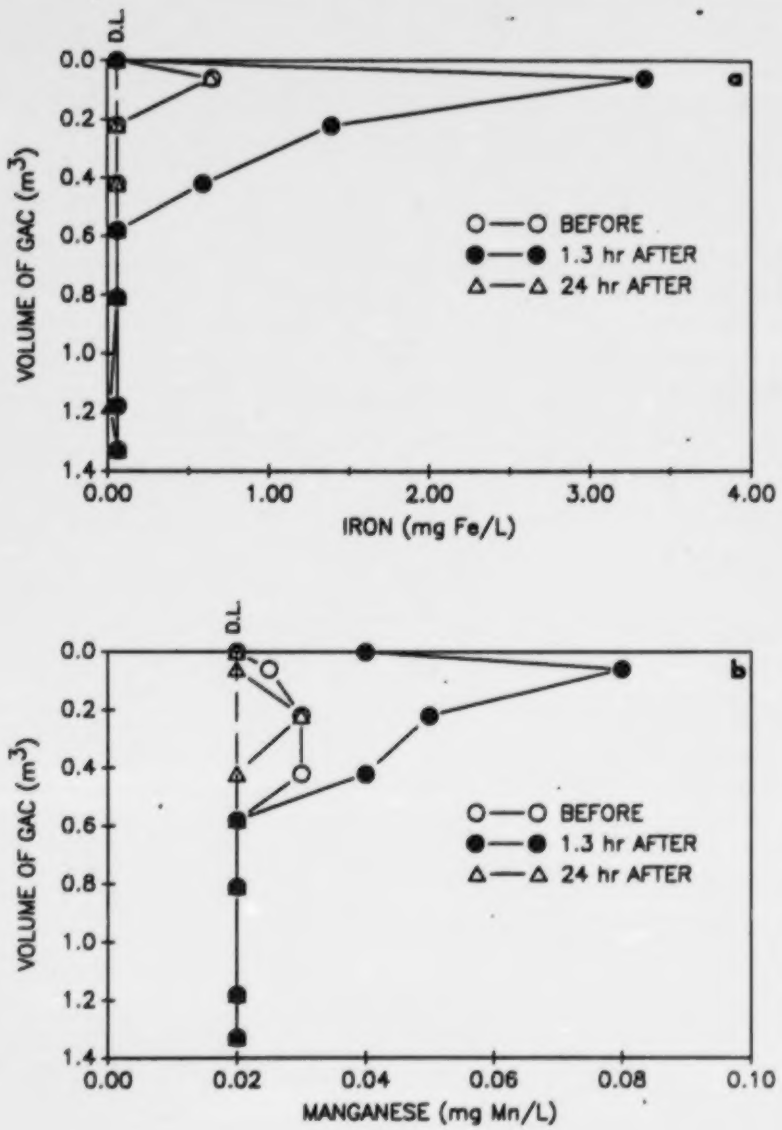


Figure 67. Backwash event - Mont Vernon, NH GAC system. Iron (a) and manganese (b) profiles before and after backwashing. (Detection limits: iron = 0.06 mg/L, manganese = 0.02 mg/L).

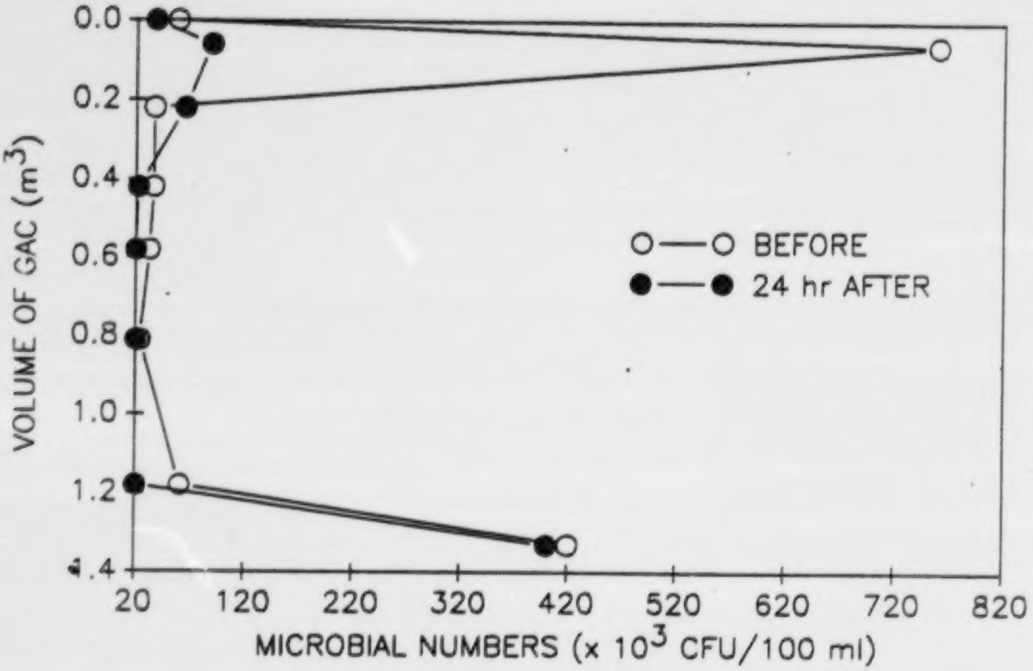


Figure 68. Backwash event - Mont Vernon, NH GAC system. Profile of microbial numbers before and after backwashing. (Detection limit = 20,000 CFU/100 mL).

The radon activity through the filter did not change as a result of the backwashing (Figure 69). There was no statistically significantly difference (α = 0.05 and 0.10, StT) in the influent and effluent radon activities obtained before or after backwashing. In a study of point-of-entry units, Lowry and Brandow (1985) determined that frequent backwashing is detrimental to radon removal because of desorption of radon from the bottom of the bed for a brief period after the backwash event. EPA (1987) noted that backwashing is detrimental to GAC point-of-entry units because it mixes the carbon in the tank. This effect was not observed in this study, however, the units were larger, there was no testing after 24 hr, and this was a single backwash event using a low backwash flowrate.

A more thorough study of backwashing needs to be done, coordinated with GAC coring before and after the backwashing events. In addition, sampling should be conducted over a longer period of time before and after backwashing to better determine the impact of the process. A complete understanding of how backwashing affects radon removal is imperative because it appears that backwashing to remove contaminants such as iron and manganese is essential to insure long term filter operation with minimal headloss. Even with pretreatment, GAC filters are usually backwashed to remove microorganisms and usage of GAC for radon removal in natural waters would probably not be an exception to this practice. In conventional practice, GAC units are backwashed with treated water, however, in small community systems this would require additional water storage adding significantly to the cost.

ECONOMICS

A detailed economic evaluation of the technologies examined in this project, for removal of radon from small community water systems, has been performed using conventional engineering economic practices. The economic evaluation assumes all community water system components (e.g., wells, storage, pumping, distribution piping) are already in-place and only the costs related to the installation and operation of the radon removal technologies are presented. Costs have been divided into two major categories: capital costs and operation and maintenance (O&M) costs. Capital costs have been further divided to include direct costs and indirect costs. Direct costs include process equipment costs plus support equipment costs (e.g., installation). All direct costs were obtained from the records of actual expenditures made during this project, hence, they are site specific. All equipment and material costs are free on board (FOB), thus, shipping costs have not been included. The indirect costs were estimated as fixed percentages of the total direct costs (TDC). These fixed percentage rates were developed by the USEPA Office of Drinking Water (Cummins, 1987) and are presented solely to provide a basis for evaluating the economic feasibility of each technology and for comparing technologies. The indirect cost estimates include: sitework (15% of TDC), engineering costs (15% of TDC), contractor overhead and profit (12% of TDC), legal and financial fees (2.5% of TDC), interest during construction (6% of TDC), and contingencies (15% of TDC).

Operation and maintenance costs include power, labor, maintenance and administrative costs. Power costs were calculated based on the equipment horsepower, operating efficiency, operation period and electrical power cost.

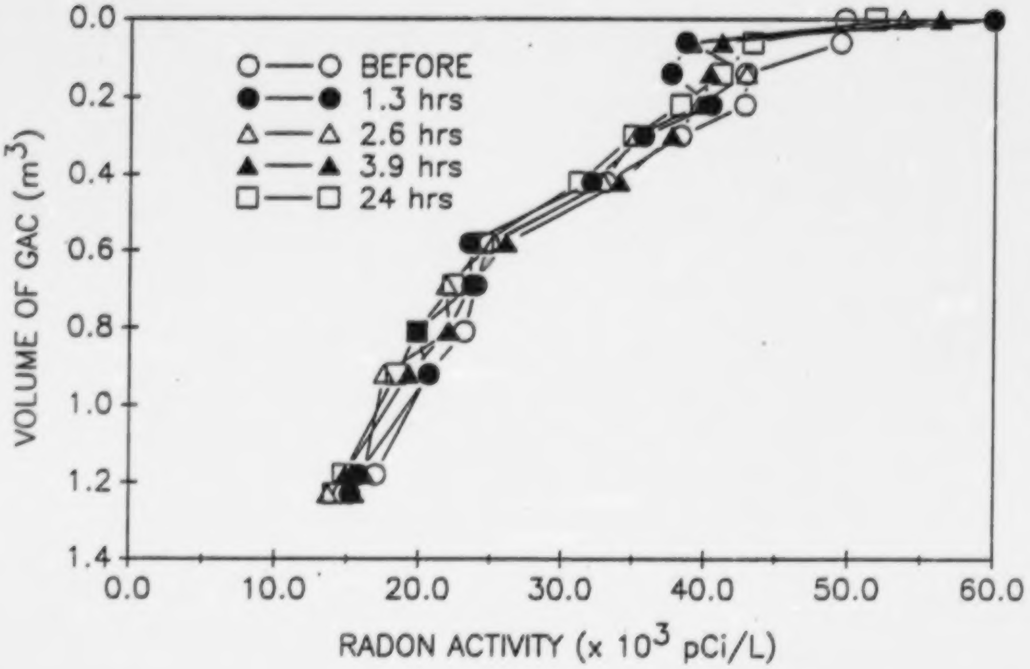


Figure 69. Backwashing event - Mont Vernon, NH GAC system. Radon activity profiles before and after backwashing.

An electrical power cost of $0.10157/kW-hr was used based on Public Service of New Hampshire (PSNH) Class G rate scale for a small community water supply with a demand of less than 5 kW (PSNH, 1989). Labor, equipment maintenance and administrative costs were calculated based on USEPA estimates (Cummins, 1987) as follows: a labor cost of $0.03/1,000 gallons was assumed; the annual equipment maintenance costs were estimated as the sum of 10% and 4% of the mechanical and non-mechanical process equipment capital costs, respectively; and the administrative costs were calculated as the sum of 20% and 25% of the labor and maintenance costs, respectively.

For comparative purposes, annual costs and production costs were developed for each treatment system. These costs were expressed in conventional units to be consistent with those reported in other studies. The annual cost was computed as the sum of the amortized capital cost and the O&M cost. The amortized capital cost is the total capital cost amortized over a 20 year time period at a 9% interest rate. A 20 year time period was chosen since it reflects a typical engineering design life for water treatment systems (Montgomery, 1985). The interest rate of 9% reflects the current (June 1989) U.S. Treasury Bill (T-Bill) interest rate. Production costs ($/1,000 gallons) were calculated by dividing the annual cost by the volume of water treated per year assuming the system was operated at the design flow.

Based on the findings of this study, that iron and manganese have the potential to foul the radon removal technologies, an additional cost for pretreatment to remove iron and manganese was estimated and used to compute the resulting production cost of radon removal with pretreatment. For comparative purposes, it was assumed that iron and manganese removal would be achieved via cation exchange. The production cost of this cation exchange system is estimated for design flows up to 38 m^3/day to be $1.50/1,000 gallons based on quotations from leading ion exchange system suppliers. This cost estimate is comparable to those reported by Reid, Lassovszky and Hathaway (1985) for use of cation exchange to remove radioactivity from drinking water. It should be noted that the cost of ion exchange for removal of iron and manganese does not include disposal of the resin regenerant or the spent ion exchange cartridges (used for non-regenerated systems). These costs may be substantial since the regenerant stream or spent cartridges are likely to contain accumulated radionuclides (e.g., uranium and radium) and require special handling and disposal. However, there is insufficient data to provide a sound basis for estimating the costs of handling and disposal of regenerant or spent cartridges. Further, it should be noted that the selection of the most cost effective pretreatment technique for iron and manganese removal is site specific and may not always involve the use of ion exchange.

All cost figures were updated to second quarter 1989 dollars using the ENR Construction Cost Index (CCI). The 1967 base year CCI index for second quarter 1989 is 426.

The cost estimates presented in this report are intended to give a general indication of the economics of the radon removal technologies studied. Specific treatment requirements will vary from site to site and may not be accurately reflected by the systems or assumptions used in this economic analysis. Further, any plans for selection, design and operation of

TABLE 10. COST ESTIMATE FOR GAC TREATMENT AT MONT VERNON, NH

Item	Total Cost (2nd Quarter 1989 dollars)[*]
CAPITAL COSTS	
DIRECT COSTS	
Process Equipment	
1 - 91 cm diameter fiberglass pressure vessel	$ 1,301
1 - 76 cm diameter fiberglass pressure vessel	905
1.33 m^3 of Barnebey Cheney Type 1002 GAC ($2091/$m^3$)	2,782
2 - PVC Distributor Systems ($98/system)	196
1 - Rockwell flow rate recorder/ totalizer	1,036
1 - Brooks Rotometer	443
1 - Differential Pressure Meter[+]	600
Total Process Equipment Cost	$ 7,263
Support Equipment	
Installation	195
Plumbing	1,545
Electrical	98
Total Support Equipment Cost	$ 1,838
TOTAL DIRECT COST	$ 9,101
INDIRECT COSTS	
Sitework	$ 1,365
Engineering	1,365
Contractor Overhead and Profit	1,092
Legal and Financial Fees	228
Interest	546
Contingencies	$ 1,365
TOTAL INDIRECT COST	$ 5,961
TOTAL CAPITAL COST	$15,062
AMORTIZED CAPITAL COST (Annual)	$ 1,650

(continued)

TABLE 10 (continued)

Item	Total Cost (2nd Quarter 1989 dollars)*
ANNUAL O&M COSTS	
Power	NA**
Maintenance	
Mechanical	208
Non-mechanical	207
Labor	71
Administrative	118
TOTAL ANNUAL O&M COST	$ 604
TOTAL ANNUAL COST	$ 2,254/year
TOTAL PRODUCTION COST (Design flow 25 m^3/day)	$0.95/1,000 gallons
PRETREATMENT COST	$1.50/1,000 gallons
TOTAL PRODUCTION COST WITH PRETREATMENT	$2.45/1,000 gallons
GAC DISPOSAL COST++	$0.19/1,000 gallons
TOTAL PRODUCTION COST WITH GAC DISPOSAL	$1.14/1,000 gallons
TOTAL PRODUCTION COST WITH PRETREATMENT AND GAC DISPOSAL	$2.64/1,000 gallons

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[*] Based on ENR Construction Cost Index, Base Year 1967 = 100.

[+] Pressure meters were not used during the research, but should be required for process control in full scale facilities.

[**] Additional power costs incurred by operation of the GAC system are considered negligible.

[++] Based on estimated disposal cost of $335/$m^3$-yr (assuming it is classified and regulated as a low level radioactive waste) and the design flow.

full-scale systems should be based on large scale, on-site pilot data reviewed by design professionals familiar with the specific site requirements.

The cost estimates for the GAC system installed in Amherst, NH are shown in Table 10. Based on the design flow of 34 m^3/day the production cost for GAC treatment at Amherst was $0.56/1,000 gallons. The production costs for this GAC system with pretreatment for iron and manganese and/or GAC disposal were also computed. The total production cost with pretreatment and GAC disposal for Amherst, NH is estimated to be $2.15/1,000 gallons.

The total annual O&M cost estimate for Amherst was $551. It should be noted that the O&M cost estimate assumes that power costs are negligible. This assumption is valid providing that the cost of the system backwashing is neglected. It is estimated that to backwash the GAC system at Amherst using a hydraulic loading rate of 4.9 m^3 H_2O/m^2-hr for a 15 min period would have a power cost of $0.09/backwashing event. Therefore, if the backwashing frequency is less often than once per day, which the data in this study indicates it should be, the cost of backwashing at Amherst would be less than $0.01/1,000 gallons.

The costs of GAC disposal, assuming it must be handled as a regulated low level radioactive waste, were computed based on an estimate (in 1989 dollars) of $4238/$m^3$ of GAC obtained from General Dynamics Services Company, Groton, CT. Based on the work of Lowry and Brandow (1985), it is assumed that with proper pretreatment and backwashing, the GAC would not require replacement or regeneration. Therefore, this disposal cost would be incurred at the end of the 20 year design life. Thus, the disposal cost was adjusted to a future cost using the ratio of ENR CCI over the previous 20 year period (1969 vs. 1989). This resulted in an estimated future cost of $17,163/$m^3$ of GAC or $14,580 for disposal of the 0.85 m^3 of GAC used in Amherst. The future cost was then translated to an annual cost assuming 9% interest over 20 years. The resulting annual cost was $335/$m^3$/yr or $285/yr. This annual cost represents an additional production cost of $0.09/1,000 gallons at Amherst.

Cost estimates for the GAC facility at Mont Vernon, NH are presented in Table 11. The costs of GAC treatment at Mont Vernon were estimated in the same manner as described for the Amherst facility. The total production cost including pretreatment and GAC disposal at Mont Vernon was estimated to be $2.64/1,000 gallons. The major differences in cost between Amherst and Mont Vernon relate to the additional equipment (i.e., pressure vessel and GAC) required to treat the higher radon loading at Mont Vernon. In particular, the additional capital and disposal costs of the GAC at Mont Vernon account for 95% of the increased costs.

The cost estimates presented for GAC treatment of radon containing well water at Amherst and Mont Vernon, NH should be interpreted with the following stipulations. (i) The costs of using GAC will be highly dependent on the influent flow and radon activity as well as the treated water radon standard which must be attained. The costs presented for Amherst are based on system design specifications of 84% radon removal at a flow of 34 m^3 and a 39,750 pCi/L average influent radon activity (0.85 m^3 GAC). Mont Vernon was designed

TABLE 11. COST ESTIMATE FOR GAC TREATMENT AT AMHERST, NH

Item	Total Cost (2nd Quarter, 1989 dollars)[*]
CAPITAL COSTS	
DIRECT COSTS	
Process Equipment	
1 - 91 cm diameter fiberglass pressure vessel	$ 1,301
0.85 m^3 of Barnebey Cheney Type 1002 GAC ($2091/$m^3$)	1,776
1 - PVC Distributor System	98
1 - Rockwell flow rate recorder/ totalizer	1,036
1 - Brooks Rotometer	543
1 - Differential Pressure Meter[+]	600
Total Process Equipment Cost	$ 5,354
Support Equipment	
Installation	215
Plumbing	1,611
Electrical	111
Total Support Equipment Cost	$ 1,937
TOTAL DIRECT COST	$ 7,291
INDIRECT COSTS	
Sitework	$ 1,094
Engineering	1,094
Contractor Overhead and Profit	875
Legal and Financial Fees	182
Interest	437
Contingencies	$ 1,094
TOTAL INDIRECT COST	$ 4,776
TOTAL CAPITAL COST	$12,067
AMORTIZED CAPITAL COST (Annual)	$ 1,322

(continued)

TABLE 11 (continued)

Item	Total Cost (2nd Quarter 1989 dollars)[*]
ANNUAL O&M COSTS	
Power	NA[**]
Maintenance	
Mechanical	218
Non-mechanical	127
Labor	100
Administrative	106
TOTAL ANNUAL O&M COST	$ 551
TOTAL ANNUAL COST	$ 1,873/year
TOTAL PRODUCTION COST (Design flow 34 m^3/day)	$0.56/1,000 gallons
PRETREATMENT COST	$1.50/1,000 gallons
TOTAL PRODUCTION COST WITH PRETREATMENT	$2.06/1,000 gallons
GAC DISPOSAL COST[++]	$0.09/1,000 gallons
TOTAL PRODUCTION COST WITH GAC DISPOSAL	$0.65/1,000 gallons
TOTAL PRODUCTION COST WITH PRETREATMENT AND GAC DISPOSAL	$2.15/1,000 gallons

[*] Based on ENR Construction Cost Index, Base Year 1967 = 100.

[+] Pressure meters were not used during the research, but should be required for process control in full scale facilities.

[**] Additional power costs incurred by operation of the GAC system are considered negligible.

[++] Based on estimated disposal cost of $335/$m^3$-yr (assuming it is classified and regulated as a low level radioactive waste) and the design flow.

for 98% radon removal at a flow of 6,500 gpd and a 155,000 pCi/L average influent radon activity (1.33 m^3 GAC). **(N.B., actual system performance did not meet the design specifications.)** Therefore, the GAC costs presented should not be used to estimate treatment costs for GAC systems with design specifications and/or performance which are significantly different than those used in this study. (ii) It is well documented in the literature for classical contaminants that the performance of GAC systems is a function of raw water quality (e.g., pH, ionic strength, anions, cations) and may vary from site to site as well as seasonally. Therefore, it is prudent to assume that the same effects should be considered for radon removal. (iii) A sensitivity analysis of the GAC cost data indicates that the cost of the system is primarily affected by the cost of the activated carbon. Since commercially available activated carbons vary in price from about \$1,413/m^3 to \$2,119/m^3, the selection of the most cost effective carbon for a specific site based on pilot studies could significantly change the system cost. (iv) The cost of GAC disposal will be dependent on state and federal regulations pertaining to the definition of and handling of low level radioactive wastes. Presently, the definition of low level wastes and hence the disposal costs vary widely from one state to another. (v) The problems of gamma emissions from GAC units removing radon must be addressed. Although it is hypothesized that these emissions can be controlled by simple, low cost shielding (e.g., water jacketing) of the units, this must be verified for small community systems. Since the technology for controlling gamma emissions has not been thoroughly developed for small communities, its costs were not included in this economic analysis. However, one should not overlook the need to design and develop costs for shielding in a full scale GAC system economic evaluation.

SECTION 5

DIFFUSED BUBBLE AERATION

INTRODUCTION

Diffused bubble aeration has been used effectively to remove many volatile contaminants from drinking water. It is easy to operate and the equipment can be retrofit into existing tanks (as long as sidewall depth effects are considered). Unlike the GAC system, it does not concentrate radon and its progeny and as a result is not a source of gamma emissions. Unlike the packed tower, it does not require a large tower structure to be built. However, in contrast to packed tower aeration, diffused bubble aeration requires contact times of several minutes or more and higher volumetric A:W ratios to achieve equivalent removals. The disadvantages most often associated with diffused bubble aeration include: high costs of operation (aeration system), release of radon to the atmosphere, potential for oxidation of iron, the need for additional pumping or repressurization of the water and excessive noise of the air blower.

Diffused bubble treatment for radon removal has been studied previously for household (Lowry and Brandow, 1981) and small community water supplies (Perkins and Hewett, 1988). The studies using small community supplies (Perkins and Hewett, 1988) were limited to an average influent radon activity of about 16,560 pCi/L and an average water flow of 0.11 m^3 H_2O/min with a volumetric A:W ratio = 10:1. For the UNH study, the average influent radon activity was 71,937 ± 8,766 pCi/L and two water flow ranges were monitored: 0.045-0.047 m^3 H_2O/min and 0.102-0.106 m^3/min at volumetric A:W ratios = 2:1, 3:1, 5:1, 7:1, 10.5:1, 15:1, and 20:1. These A:W ratios and water flowrates were chosen to determine the effect of air and water flowrates on radon removal efficiency. Costs, the effects of water quality, such as iron and turbidity, and stack gas emissions were also considered during the diffused bubble study.

METHODS AND MATERIALS

Description of Units

The diffused bubble aeration system, designed by Dr. Jerry Lowry (Unity, ME), consisted of 3 polyethylene tanks aligned in series, each with a holding capacity of 1022 L (Figure 70). The system was installed in a small community water supply in Derry, NH. An insulated, plywood building protected the system from adverse weather and vandalism. Aeration was provided by an air blower with a 2.66 m^3/min capacity which forced ambient outdoor air into the tanks via Schedule 40, 3.8 cm diameter PVC piping connected to the diffusers

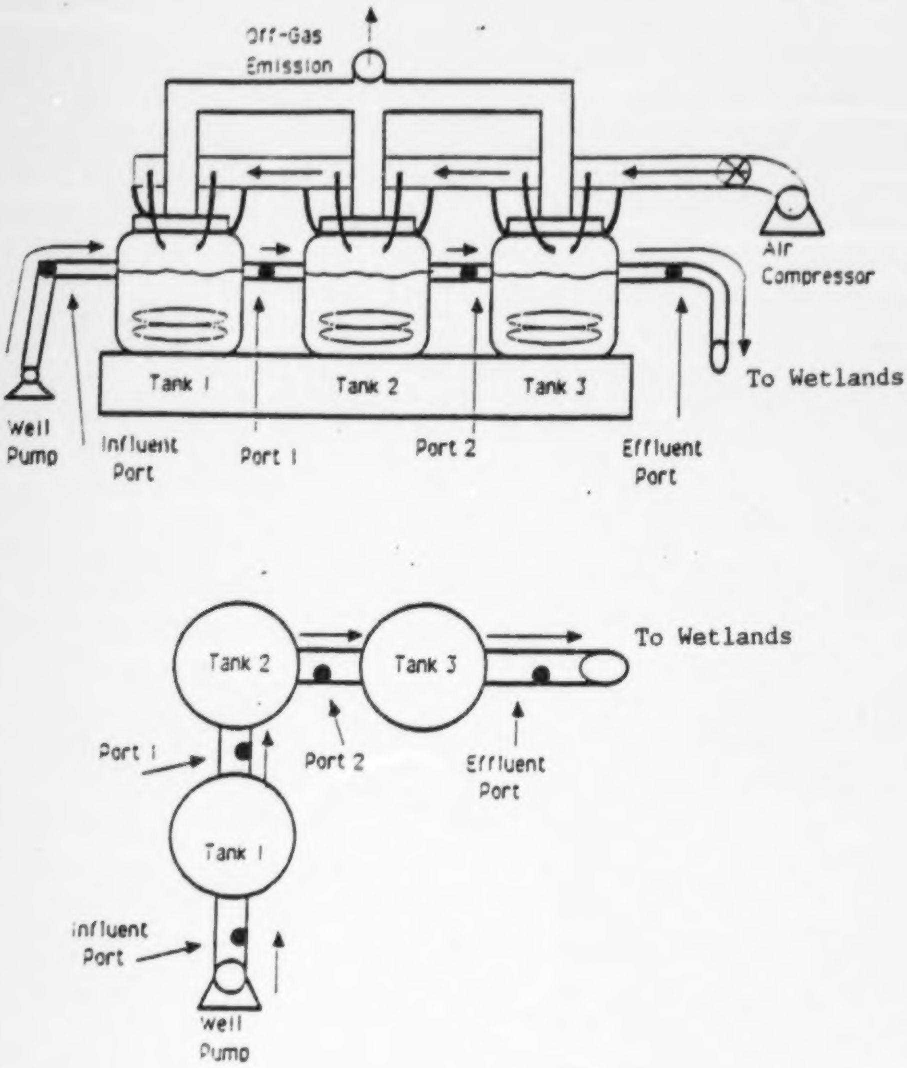
Off-Gas
Emission
Air
Compressor
To Wetlands
Tank 1
Tank 2
Tank 3
Well
Pump
Influent
Port
Port 1
Port 2
Effluent
Port
Tank 2
Tank 3
To Wetlands
Port 1
Port 2
Effluent
Port
Tank 1
Influent
Port
Well
Pump

Figure 70. Schematic of the diffused bubble aeration system. Derry, NH.

which consisted of 1.90 cm diameter coiled plastic tubes with 0.038 cm holes drilled in their underside. The distance between the holes ranged from 0.5 to 1.6 cm. Air flow was regulated with a globe valve located on the influent air line. The diffusers were located about 79 cm below the water's surface and 36 cm from the bottom of each tank. Once the radon was stripped from the water, the contaminated air flowed from the top of each tank through Schedule 40 3.18 cm PVC piping and was vented outside of the building.

Water was pumped directly from the bedrock wells into the system through 3.18 cm plastic hose. The water from each upstream tank overflowed into the adjacent downstream tank by simple gravity displacement. Since the water supply was not being used by the community, treated water bypassed the atmospheric storage tank and flowed from the third tank by gravity through a Schedule 40, 5.1 cm PVC pipe to a wetland area outside of the pumphouse. The first two tanks maintained higher water volumes, of about 38 liters due to the expansion of the water column by the air.

An influent water sampling port was located in the influent line to the first tank (Figure 70). The 5.1 cm PVC connections between Tanks 1 and 2 and Tanks 2 and 3 were tapped using ball valve faucets with 1.1 cm diameter hose connections to provide sampling ports. The hoses attached to the ports each had the neck of a polyethylene funnel (10 - 15 cm maximum opening) fitted into their free end. During sampling, the mouth of the funnel was upright so that the water from the port filled it and flowed over its lip. In this way a radon sample could easily be collected from the funnel with little disturbance at a low water flowrate. An effluent sampling port was similarly constructed and tapped into the 5.1 cm PVC pipe carrying effluent water from Tank 3 to the wetland. The PVC connections between tanks were located 114 cm above the bottom of each tank.

Experimental Design

The system allowed comparison of the removal efficiencies over a wide range of influent radon activities (Tank 1: 60,843 to 86,355 pCi/L radon, Tank 2: 10,096 to 80,271 pCi/L and Tank 3: 1,767 to 74,112 pCi/L). Two flow ranges were obtained by manually operating one (low flow = 0.047 ± 0.00053 m^3 H_2O/min) or two wells (high flow = 0.10 ± 0.0019 m^3/min). Radon activities averaged 65,487 ± 5,657 pCi/L during high water flow and 78,385 ± 6,120 pCi/L during low water flow. The two water flowrates and the two radon activities resulted in applied radon loading rates averaging 6,819 ± 548 nanoCuries (10^3 pCi) per minute (nCi/min) for high water flow and 3,639 ± 295 nCi/min for low water flow. A:W ratios of 2:1, 3:1, 5:1, 7:1, 10.5:1, 15:1, and 20:1 were tested for both water flowrates. The tanks were drained at the end of each run and refilled with raw water immediately before the start of a new run.

Sampling Plan

The parameters monitored for the diffused bubble aeration system are shown in Table 12. Samples were collected at 0, 15, 30, 60 and 90 min at both water flowrates and also at 120 and 150 min for the low flowrate. Water flowrate was monitored using an in-line water flowmeter/totalizer located in

TABLE 12. DIFFUSED BUBBLE AERATION SAMPLING PARAMETERS

	Radon	Alkalinity	Turbidity	Temperature	Iron/Mang.*	Flovrate
Water Samples						
- Influent	X	X	X	X	X	X
- Port 1	X					
- Port 2	X					
- Effluent	X	X	X	X	X	X
Air Samples						
- Influent						X
- Building				X		
- Effluent	X					

---

[*] Iron and manganese.

the influent piping directly before the diffused bubble system. Air flowrate was monitored using a pitot tube installed in the influent air pipe and connected to a Dwyer^R Magnehelic pressure difference gauge. Air samples for radon analysis were taken before each run (background) and approximately 1 hr into the run. A 0.97 cm diameter plastic tube, which was placed 20 cm inside the off-gas (air effluent) pipe, was used to pump air samples to a Pylon AB-5 monitor.

RESULTS AND DISCUSSION

Radon

Overall System Performance--

Figures 71 and 72 show percent removals of applied radon over time for each run (A:W ratio and high or low water flowrate combination). Percent removals were calculated by dividing the mass of radon removed per time (i.e., applied radon loading ($Q_w \times C_{inf}$) - radon remaining ($Q_w \times C_{eff}$)) by the applied radon mass loading ($Q_w \times C_{inf}$) and multiplying by 100. (This is equivalent to $(C_{inf} - C_{eff})/C_{inf}$ because Q_w factors out of the equation.) A mean influent radon activity was computed for each run and used in the percent removal calculation. This approach is valid since there was <15% variation in influent activity during a run.

ANOVA analyses were run on the data to determine when the system was at steady state for each run. This determination was necessary because the three tanks in the diffused bubble system were filled with raw water at the beginning of a run. Steady state was defined as the minimum time after which effluent radon activities at a given condition (A:W ratio and Q_w) were not significantly different at $\alpha = 0.05$. However, true steady-state conditions could not be achieved because there were changes in the raw water quality and water flowrate from the wells over time. Table 13 shows the time required to reach steady state removal at all conditions tested. The times required for the high water flowrate conditions were shorter than those for the lower flowrate. For all subsequent statistical analysis of the diffused bubble system, different conditions were compared using data collected at 60 and 90 min for high flow and 90, 120 and 150 min for low flow. These times were chosen because for all A:W ratios tested the radon removals were at steady state.

For a diffused bubble system of a given construction, the percent removal of the radon applied is controlled by (i) the contact time between the contaminated water and diffused air (the hydraulic detention time), (ii) the aeration system's K_La (which is temperature dependent), (iii) the concentration of the contaminant in the air, and (iv) the influent concentration of the contaminant in the water.

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*Q_w = water flowrate (volume/time)

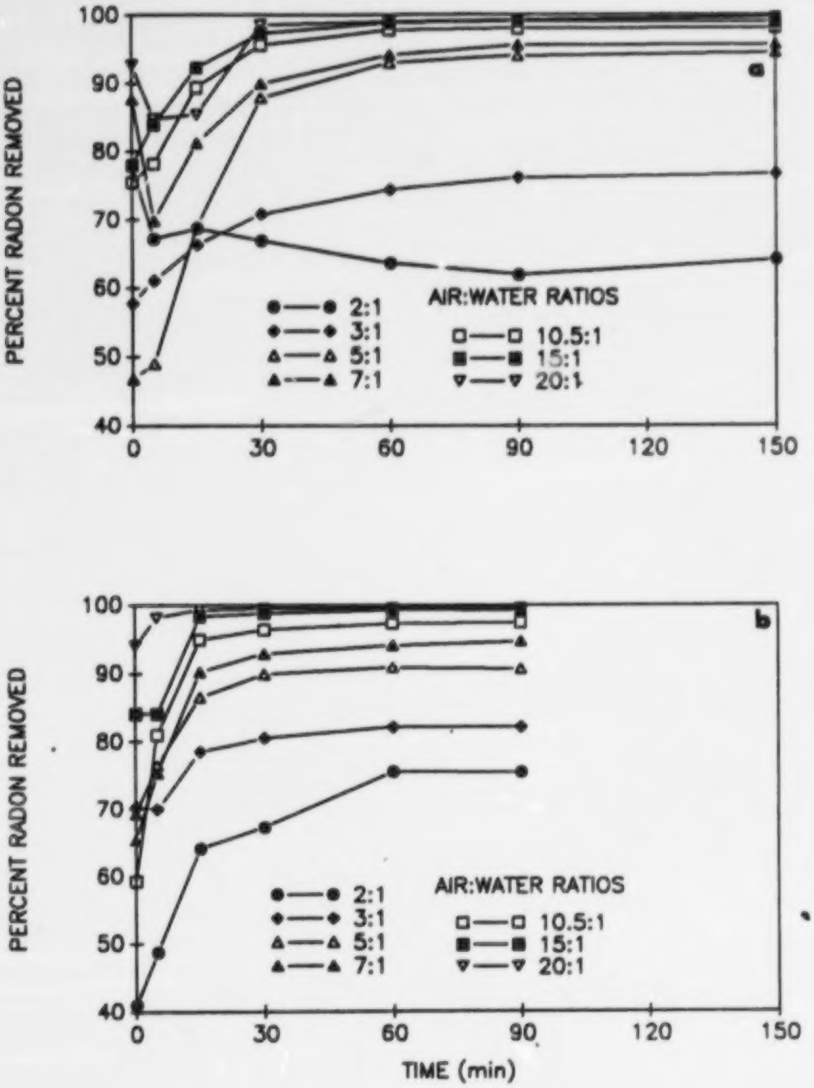

Figure 71. Diffused bubble aeration - Derry, NH. Percent radon removal as a function of time for varying A:W ratios.

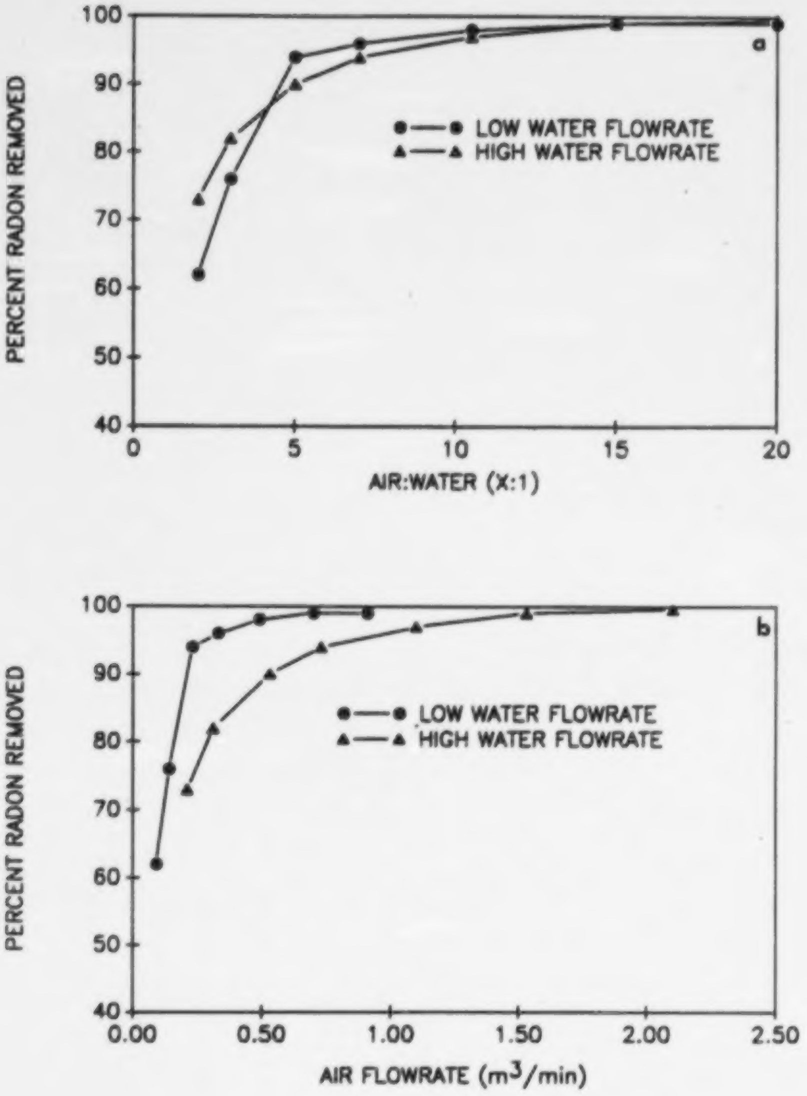

Figure 72. Diffused bubble aeration - Derry, NH. Percent radon removal as a function of A:W ratios (a) and air flowrate (b) for the low and high water flowrate.

TABLE 13. TIME REQUIRED TO ACHIEVE STEADY STATE OPERATION* DURING THE DIFFUSED BUBBLE EVALUATIONS

A:W Ratio	Time (min) Low Water Flowrate	Time (min) High Water Flowrate
2:1	60	60
3:1	90	60
5:1	90	60
7:1	90	60+
10.5:1	60	60
15:1	30	15
20:1	30	15

[*] Minimum time after which there was no significant difference in effluent quality achieved in diffused bubble system, $\alpha = 0.05$ (ANOVA).

[+] Not significantly different at $\alpha = 0.04$, significant different at $\alpha = 0.05$.

Figure 71 shows the overall percent removals of radon applied vs. A:W ratio for the high and low water flowrates using data after steady state conditions were achieved. As the A:W ratio increased from 2:1 to 5:1 for each flowrate, there was a sharp increase in radon removal. However, above 5:1, there was much less improvement in efficiency with large increases in A:W ratio.

When operating at A:W ratios of 5:1 and greater (at both high and low water flowrates), the overall radon removal efficiency ranged from 90.0% to >99.6%; the greatest efficiency was obtained at A:W ratios of 15:1 and 20:1. At A:W ratios of 10.5:1 and greater for the low flowrate and 15:1 and greater for the high flowrate, there was no significant difference in removal efficiency ($\alpha = 0.05$ and 0.01, ANOVA). Hence, for the diffused bubble system tested at the given conditions of radon loading, the lowest A:W ratios to yield (statistically significant) maximum radon removal for low and high water flowrates were in the range 7:1 to 10.5:1 and 10.5:1 to 15:1, respectively. From the practical viewpoint of designing the diffused bubble system, the A:W ratio should be based on the most cost effective blower size and mode of operation. Given the design water flowrate of 37.85 m^3/day and assuming a minimum A:W of 20:1 (0.53 m^3/min) with an estimated static pressure of 38 cm-H_2O, the most cost effective (capital and power rating) input horsepower size for the blower would be 0.5 hp (R.G. Wilson; Aladdin HVAC, Corp; personal communication; 1989). Using this blower represents a conservative engineering design (i.e., a 1.13 m^3/min air flow is delivered) since the resulting A:W ratio would be 43:1. The data suggests such a high A:W ratio would achieve removals of $\geq$99.6% with an influent averaging 72,000 pCi/L.

At the lowest A:W ratio (2:1) at both water flowrates, the low air flow (<0.23 m^3/min) severely limited bubble distribution in the tanks, resulting in lower radon removal (Figure 73). At air flowrates of <1.0 m^3/min, the percent removal achieved with the low water flowrate was significantly greater than the higher water flowrate in spite of the significantly higher radon activities entering the tank during the low flow runs (High = 65,487 ± 5,657 pCi/L vs. Low = 78,385 ± 6,120 pCi/L) ($\alpha = 0.05$ and 0.01, StT). This difference may have been a function of the longer detention time in the units at the low water flowrate condition. The overall mass removal of radon (pCi/min) was, however, considerably greater at the higher water flowrates for comparable air flowrates (Figure 73). These results reflect the greater mass loading ($Q_w \times C_{inf}$) of radon to the units which negated the effect of the decreased detention time by increasing the rate of mass transfer (i.e., higher concentration gradient $C_{liquid} - C_{gas}$) particularly in Tanks 2 and 3.

Previous studies of diffused bubble aeration performance on removal of radon are comparable to those obtained with the system at Derry. Perkins and Hewett (1988) found that a diffused bubble system employing 61 cm diameter stainless steel bowl diffusers containing fritted plastic diffuser media could achieve percent removals of radon activities of 95% and greater. Their system was run at an A:W = 10:1, and a 0.23 m^3 H_2O/min water flow, with a hydraulic detention time of 60 min and a raw water radon activity of 16,560 pCi/L. Their setup consisted of 2 manholes, 2.4 m in diameter and 2.4 m deep so that

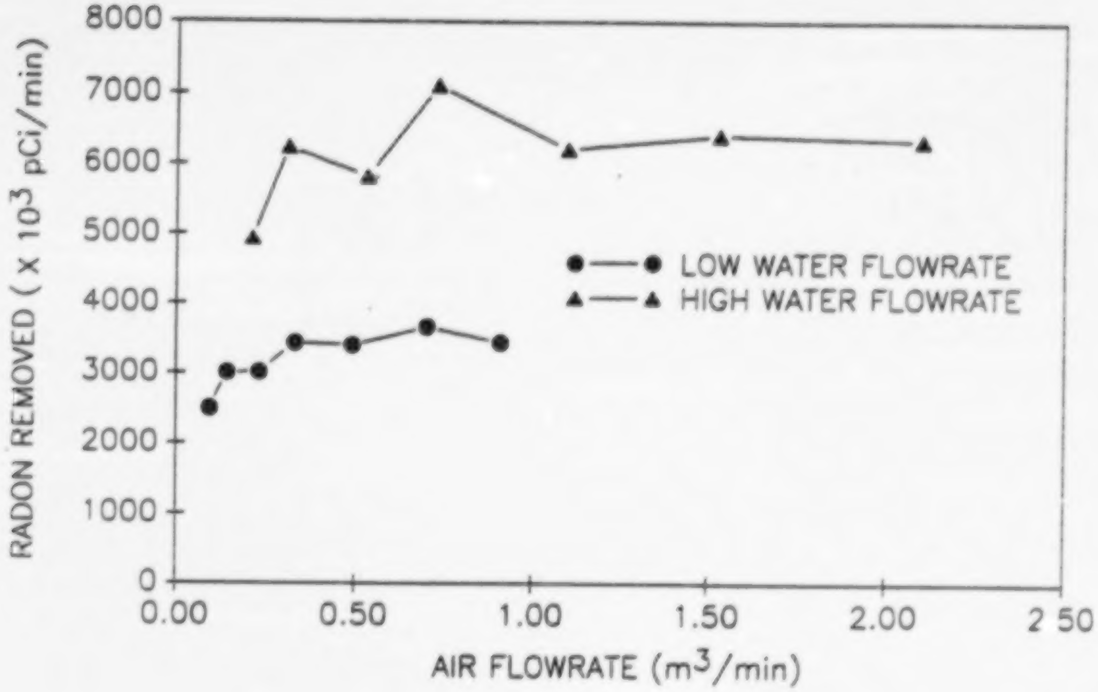

Figure 73. Diffused bubble aeration - Derry, NH. Radon activity removed (x 10^3 pCi/min) as a function of an air flowrate for the low and high water flowrates.

sidewall depth was more of a factor than in the Derry system. A third manhole, 1.8 m in diameter and 2.4 m deep, acted as a clear well.

Lowry and Brandow (1981) also found that 95% removal of radon activity could be achieved with diffused aeration. The system they tested was a static (Q_w = 0 m^3/min) tank (132 - 151 L), usable in point-of-entry applications. The influent radon activity was 30,000 pCi/L, and yielded an effluent activity of 1,100 pCi/L with a detention time of 30 min and an air flow of 0.014 m^3/min. They estimated that 95-100% removal of applied radon activity could be achieved for a continuous flow system after 60 min of operation.

In more recent research, Lowry et al. (1987) tested a 454L vessel with fine bubble diffusers using an aeration rate of 1,400 L/hr and attained 99% removal with a 60 min aeration period (A:W ratio = 3.4:1). A multi-stage bubble aeration system (Lowry et al., 1988) has been installed in 22 point-of-entry applications and has achieved radon removals of 92 to 99.9% with influents ranging from 20,000 pCi/L to over 500,000 pCi/L.

Though results from all of these diffused bubble systems are similar to the Derry results, extrapolating from individual system performance at a given loading and with different construction (including diffusers) and water quality to other water supplies, especially those with considerably lower radon mass loadings, should be avoided because of variations in mass transfer characteristics.

Individual Tank Performance--

The performance of the individual tanks was analyzed to better understand what was occurring within the diffused bubble system. It was designed to act as three completely stirred tank reactors in series. In initial testing of the diffused bubble system at the Derry site, the results indicated that there were problems with the construction (Figure 74). Tank 2 removed ≤18% of the total radon regardless of A:W ratio or water flowrate. In addition, at the lower A:W ratios (2:1, 3:1, 5:1), Tank 1 was also not performing well. Visual observation indicated that air flow was somewhat restricted to Tank 2, but this was hardly noticeable. Dye studies indicated that hydraulically all tanks were completely mixed and not short circuiting even at the lowest A:W ratios and high water flowrate. The theoretical detention time of 9.6 min for each 1022 L tank at the high water flowrate of 0.11 m^3 H_2O/min showed excellent agreement with the actual detention times shown in Figure 75. The air supply lines to all tanks were moved to a single header, which improved the removal in Tank 1, but had no effect on Tank 2. Upon careful examination of the system, the brackets holding the diffuser in Tank 2 were found to be 7.6 cm lower on one side of the circular tank than on the other. Tank 1 was also raised 1.0 cm to be level with Tanks 2 and 3. These improvements yielded markedly better performance in Tank 2, however, equal air distribution to Tank 1 continued to be a problem at air flows <0.23 m^3/min. These data indicate that equal tank and diffuser elevations (i.e., equal air distribution) in small community multiple tank systems are crucial in achieving efficient radon removal. Further, diffusers must be operated at air flowrates within their design specifications.

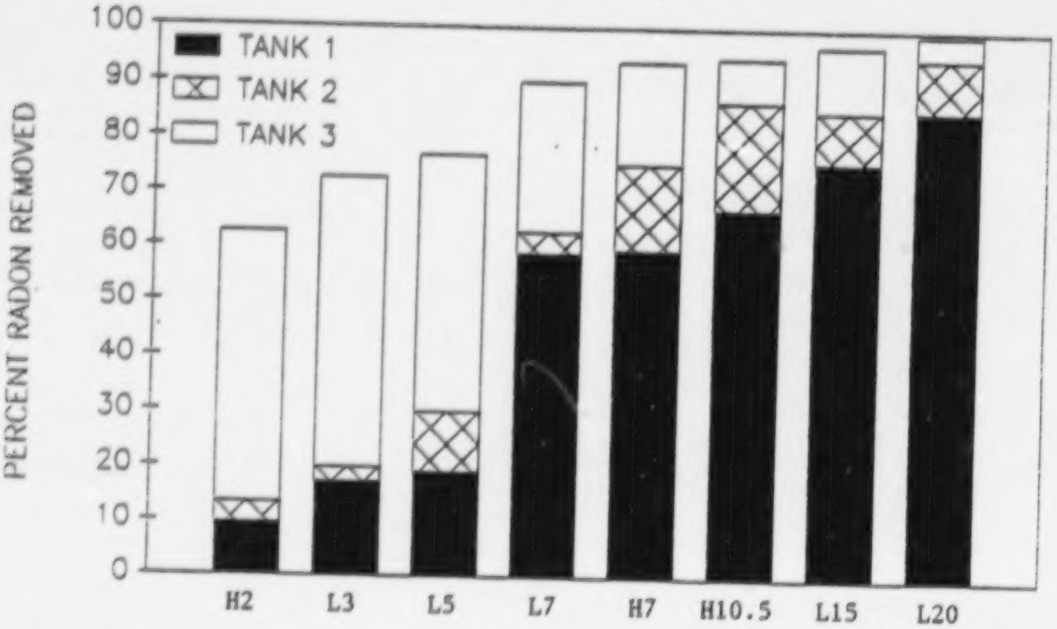

Figure 74. Diffused bubble aeration - Derry, NH. (Before repairs to aeration systems). Percent radon removal for high (H) and low (L) water flowrates and varying A:W ratios in each tank.

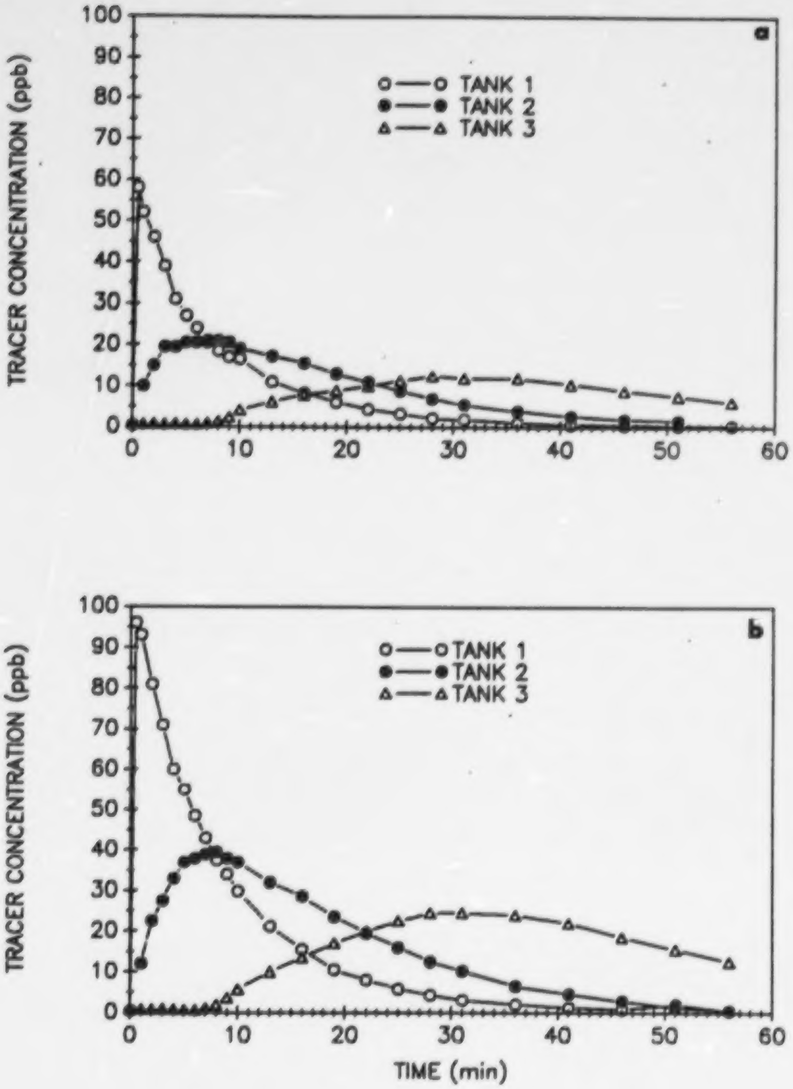

Figure 75. Diffused bubble aeration - Derry, NH. Dye concentration as a function of time in each tank. (a) water flowrate = 0.106 m³/min, A:W ratio = 3:1. (b) water flowrate = 0.106 m³/min, A:W ratio = 10.5:1.

After these problems were corrected, the diffused bubble system yielded the results shown in Figure 76. At the lowest A:W ratio, the percent radon removal in the third tank was greater than in the first or second tanks indicating the continued difficulty in getting equal air distribution in a small system unless the construction details are more sophisticated. As the A:W ratio increased, the efficiency in the first tank increased from 39% at A:W = 5:1 (high water flowrate) to 85% at A:W = 20:1 (high water flowrate). As air flowrates increased, the radon removal observed in Tank 1 increased (Figures 76 and 77). However, as the removal increased in the first tank, Tanks 2 and 3 became increasingly less efficient with respect to the overall process and served as polishing units. The data suggest that for the diffused bubble system tested at water flowrates of 0.047 and 0.10 m^3 H_2O/min radon removal was highly dependent on the radon activity and therefore the concentration entering the tank.

Radon removal in the diffused bubble system appeared to be a function of mass transfer. As the radon activity became progressively lower through the series of tanks, the driving force ($C_{liquid} - C_{gas}$) decreased, limiting removal. It is also possible that in some applications where radon activity is very low in the water, the activity in the air could be significant, making gas phase mass transfer limited. Similarly, if the radon concentration in the air above the tanks is allowed to accumulate (i.e., poor exhaust gas venting), transfer limitations may also occur. Though mass transfer may make it more difficult to achieve low effluent activities, the diffused bubble system tested produced water with effluent activities of about 1,849 to 280 pCi/L for A:W ratios of 10.5:1 and greater (Table 14). These data suggest that it may be possible to meet an MCL of 200 pCi/L if the air flowrate is high, and/or there is a long contact time.

Radon removal in diffused bubble systems is dependent on many factors not evaluated in this study including: sidewall depth, bubble size and diffuser type, air and water temperature, and tank and diffuser configuration. Though it appears that most diffused bubble systems tested (Perkins and Hewett, 1988; Lowry and Brandow, 1981 and this study) can achieve high percent radon removal, pilot scale testing of a given configuration may be required especially when (i) retrofitting diffusers into existing tanks in a small community water system, (ii) treating water with low influent radon activity and/or (iii) attempting to meet low MCLs.

Mass Transfer Coefficients--

Theoretically, the global liquid mass transfer coefficient, K_La, for a given diffused bubble system is primarily a function of the air flowrate (Q_g) which can affect the bubble diameter (d_b), the contact time as the bubble rises which is a function of sidewall depth (H_L) and bubble velocity, the water characteristics such as viscosity (μ_w) and density (ρ_w), the contaminant diffusivity compared with oxygen (β), oxygen diffusivity in water (D_o), and

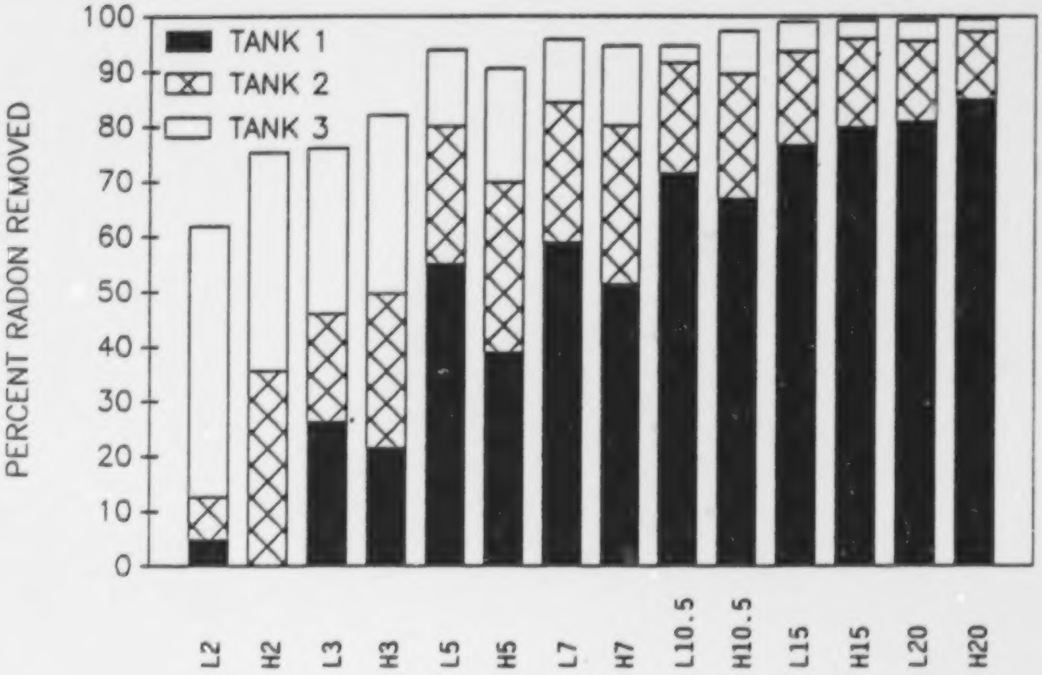

Figure 76. Diffused aeration - Derry, NH. (After improvements to aeration system). Percent radon removal as a function of high (H) and low (L) water flovrates and varying A:V ratios in each tank.

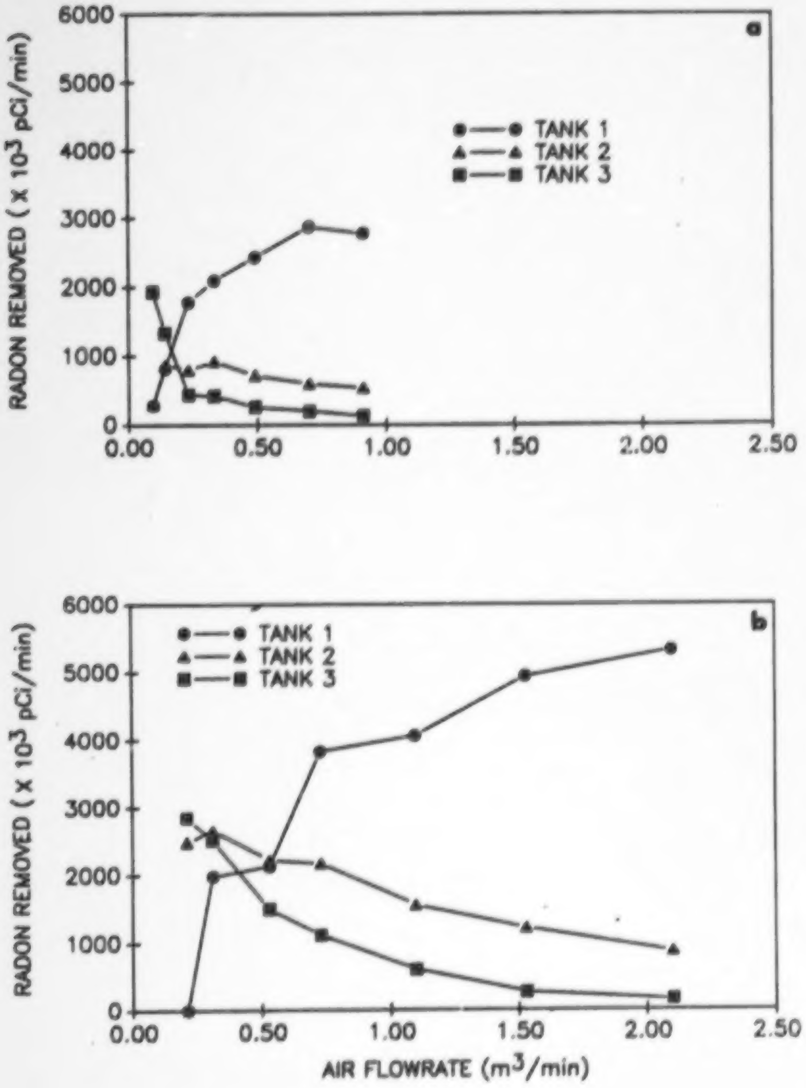

Figure 77. Diffused bubble aeration - Derry, NH. Radon activity removed (x 10^3 pCi/min) as a function of air flowrate for low (a) and high (b) water flowrates in each tank.

TABLE 14. EFFLUENT RADON ACTIVITIES FROM THE DIFFUSED BUBBLE SYSTEM OPERATING AT A:W RATIOS OF 10.5:1 AND GREATER

Water Flowrate	A:W Ratio	Radon Activity (pCi/L) Influent[*]	Effluent
Low	10.5:1	74,986 ± 5,894	1,541 ± 87
	15:1	79,799 ± 2,878	777 ± 16
	20:1	76,609 ± 4,995	556 ± 49
High	10.5:1	61,352 ± 7,406	1,849 ± 48
	15:1	69,392 ± 6,003	564 ± 73
	20:1	60,843 ± 4,729	280 ± 14

[*] Raw water radon activity from wells to diffused bubble system.

the water temperature (since the water density, viscosity, and diffusivity are all temperature dependent). The K_La can be expressed as:

$$K_La = \frac{6\beta' \, H_L^{2/3} Q_g \rho_v^{1/2} D_o^{1/2}}{d_b \, V_L \, (\mu_v)^{1/2}} \qquad \text{(eq. 5)}$$

where $\beta' = \beta / H_L^{1/3}$ and V_L = the liquid volume of the tank (Schroeder, 1977).

For a given diffused bubble system, assuming well mixed conditions, the fraction of the radon removed ($1 - C_{eff}/C_{inf}$) can be estimated using the following steady state formula (Eckenfelder,1989):

$$1 - \frac{C_{eff}}{C_{inf}} = 1 - \left[1 + \frac{Q_g}{Q_L} H_c \, (1 - e^{-\phi})\right]^{-1} \qquad \text{(eq. 6)}$$

where C_{eff} = effluent radon activity in water at steady state, C_{inf} = raw water radon activity, Q_g = air flowrate, Q_L = liquid flowrate, H_c = dimensionless Henry's constant, and ϕ = bubble saturation parameter defined as:

$$\phi = \frac{K_La \, V_L}{H_c \, Q_g} \qquad \text{(eq. 7)}$$

The ϕ term accounts for the fractional saturation of the air bubble with respect to radon.

Figure 78 shows the relationship between air flowrate and the fraction of radon removed for each tank at high and low water flowrates. Theoretically, the fraction removed for each tank (assuming identical design and operation) should be the same (eq. 6 and 7). However, Tank 3 consistently had significantly higher removals ($\alpha = 0.01$, StT) than Tanks 1 and 2 under all conditions except the A:W ratio of 20:1 for the high water flowrate (Table 15). Further, Tank 2 had significantly ($\alpha = 0.01$, StT) higher removals than Tank 1 in most cases. These observed differences may have been a function of the assumption that all tanks received an equal air flowrate. The construction and operation of the air distribution system appeared to favor Tank 3, with Tank 1 receiving the least flow. The decreased air flowrate to Tank 2 and especially to Tank 1 would account (eq. 6 and 7) for the decreased removals observed in those tanks. As predicted by eq. 6 and 7, the removals increased significantly ($\alpha = 0.05$, StT) as air flowrate increased in all cases except high water flowrate (A:W = 15:1 and 20:1) and low water flowrate (A:W = 2:1 and 15:1 and 20:1) which were only different at $\alpha \geq 0.10$ (StT).

Differences in tank volume, diffuser type and configuration, air flowrate, sidewall depth, water quality and temperature make comparisons of K_La values (eq. 5) and removals (eq. 6 and 7) among diffused bubble systems

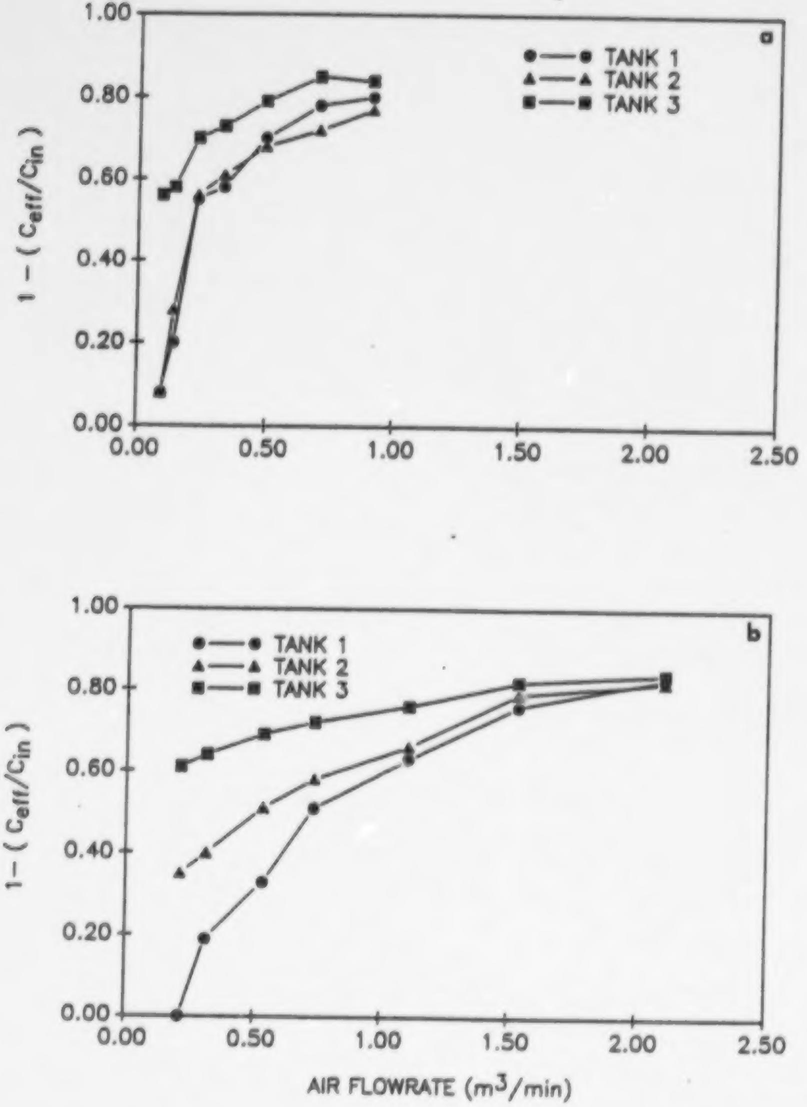

Figure 78. Diffused bubble aeration - Derry, NH. Fraction of radon removed ($1-C_{eff}/C_{inf}$) as a function of air flowrate for low (a) and high (b) water flowrates in each tank.

TABLE 15. STATISTICAL COMPARISONS[*] OF FRACTION OF RADON REMOVED $(1 - \frac{C_{eff}}{C_{inf}})$ IN TANKS IN THE DIFFUSED BUBBLE SYSTEM

Water Flowrate	A:V Ratio	Statistical Relationship	Significance Level α
Low	2:1, 5:1, 10.5:1	Tank 1 = Tank 2	0.05 & 0.10
	3:1, 7:1, 15:1, 20:1	Tank 2 > Tank 1	0.05 & 0.01
	2:1, 3:1, 5:1, 7:1, 10.5:1, 15:1, 20:1	Tank 3 > Tank 1	0.05 & 0.01
		Tank 3 > Tank 2	
High[+]	3:1, 5:1, 7:1, 10.5:1	Tank 2 > Tank 1	0.05 & 0.01
		Tank 3 > Tank 1	
		Tank 3 > Tank 2	
	15:1	Tank 2 > Tank 1	0.05 & 0.01
		Tank 3 > Tank 1	0.05 & 0.01
		Tank 3 > Tank 2	0.05
		Tank 3 = Tank 2	0.01
	20:1	Tank 1 = Tank 2	0.05 & 0.10
		Tank 3 = Tank 1	0.05 & 0.10
		Tank 3 > Tank 2	0.05
		Tank 3 = Tank 2	0.01

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[*] As assessed by ANOVA.

[+] High water flowrate and A:V ratio of 2:1 not included because of air flow distribution problems in Tank 1.

inappropriate (Eckenfelder, 1989; Perkins and Hewett, 1988; Lowry and Brandow, 1981).

Off-Gas Radon Activities--

Stack emissions were monitored to determine if the off-gas radon activities from the diffused bubble system would affect the air quality of the surrounding environment. The measured off-gas radon emission rates were plotted against the theoretical off-gas radon emissions rates (Figure 79). Theoretical off-gas rates were found by multiplying the actual difference between the measured influent and effluent water radon activities in the diffused bubble system by the measured air flowrates for a given operating condition. The measured activities were 10% to 50% lower than those predicted by theoretical calculation. The differences may have been a function of (i) variation in radon loading over the runs, (ii) lack of replicate sampling, (iii) slight variations in the air flowrate, (iv) dilution of the sample collected with ambient air at the end of the stack and/or (v) air sampling problems from streamlining of the off-gas flow around the sampling tube.

As the A:W ratio and air flowrate increased, there was a decrease in the radon activity in the off-gas (Figure 80). The increased air flow diluted the mass of radon removed from the water in increasingly larger volumes of air. Higher air flowrates would be expected to decrease the off-gas activity even further. For the system tested, the off-gas activity (3,361 to 18,356 pCi/L) would need to be diluted 10^4 to 10^5 times to be similar to radon activities found in the ambient air at the site (0.1 to 0.15 pCi/L) (NH Bureau of Radiological Health, personal communication, 1986).

Iron and Manganese

As observed with radon, there was a difference in the influent total iron and manganese concentrations in the raw water between the high and low water flowrate tests (Low Water Flowrate: 0.14 ± 0.11 mg Fe/L and 0.10 ± 0.05 mg Mn/L; High Water Flowrate: 0.31 ± 0.28 mg Fe/L and 0.32 ± 0.04 mg Mn/L). This probably occurred because of differences in the wells over time at the Derry site. In all cases, except low water flowrate A:W = 3:1 (L3:1), there was no significant change in total iron through the diffused bubble system ($\alpha = 0.05$ and 0.10, StT). At L3:1, the influent iron 0.31 ± 0.01 mg/L decreased to below detection (0.06 mg/L) in the effluent. In all cases, except for high water flowrate A:W = 3:1 (H3:1), there was also no significant change ($\alpha = 0.05$ and 0.10, StT) in total manganese through the system. At H3:1 the influent manganese decreased from 0.30 ± 0.14 mg/L in the influent to 0.13 ± 0.04 mg/L in the effluent. Overall, there was little change in the total concentration of either metal through the system. The effects of aeration on metal oxidation would not be noticeable in a total metals analysis unless settling were occurring in the completely mixed diffused bubble system, which is doubtful. In the future, soluble and total metal concentrations should be analyzed to determine if oxidation and precipitate formation are factors. This would be problematic if it occurred because of potential accumulation of sludges in the atmospheric storage tank and fouling of the diffusers.

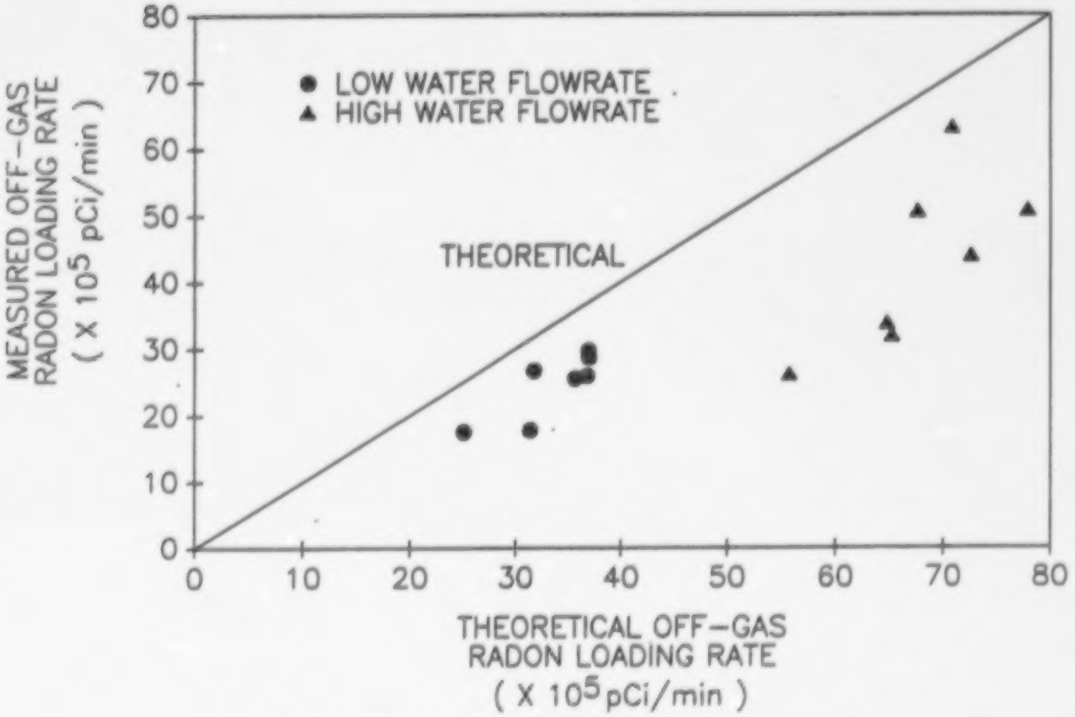


Figure 79. Diffused bubble aeration - Derry, NH. Off-gas radon activity (x 10^5 pCi/min) as a function of theoretical off-gas radon activity (x 10^5 pCi/min).

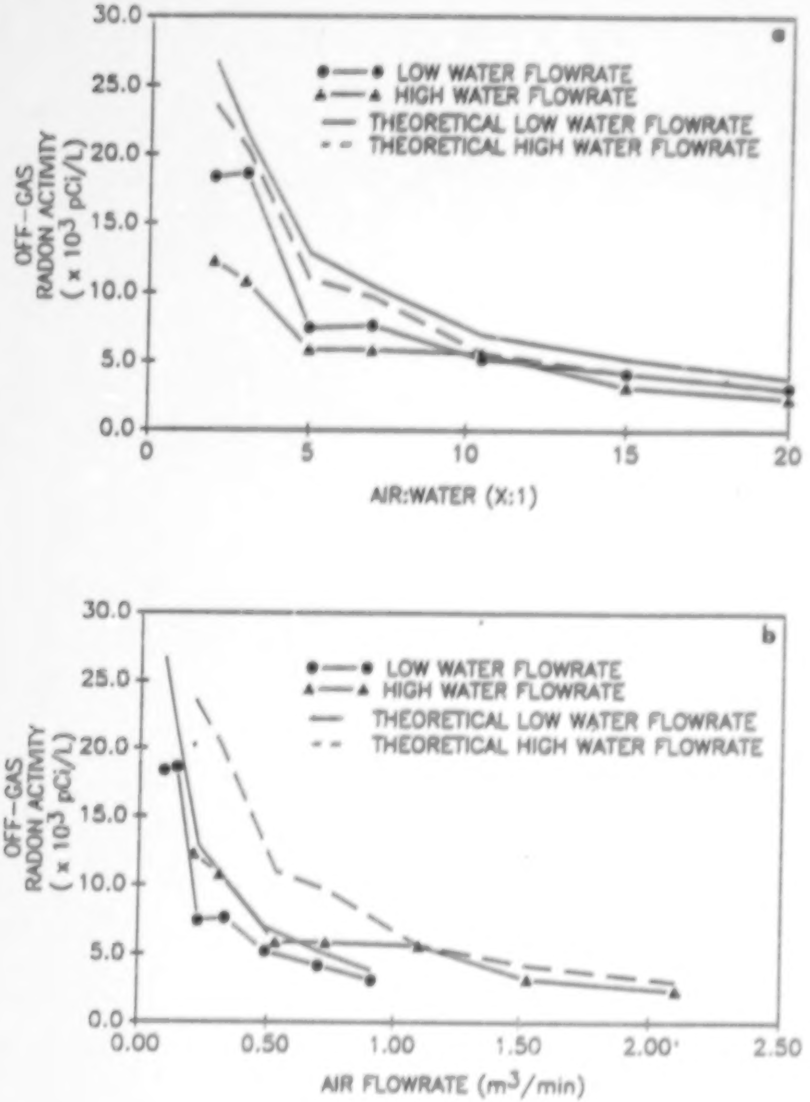


Figure 80. Diffused bubble aeration - Derry, NH. Off gas radon activity as a function of A:W ratio (a) and air flowrate (b) for the low and high water flowrates.

Turbidity, Alkalinity and pH

The influent turbidity averaged 0.20 ± 0.09 NTU and 0.08 + 0.07 NTU for the high and low water flowrates, respectively. With the exception of the low water flowrate at an A:W ratio of 2:1, there was no significant difference in turbidity through the units at all A:W ratios and low water flowrate (α = 0.05 and 0.10, StT). At the high water flowrate for A:W ratio 2:1 to 5:1, there was no significant change in turbidity through the units (α = 0.05 and 0.01, StT). However at A:W ratios of 7:1 and greater significant increases in turbidity occurred (Table 16). It appears that at these air flowrates, there may have been some increase in precipitate formation within the units, but this cannot be corroborated with the iron data because the latter does not reflect changes in soluble iron. It is unlikely that manganese precipitates would have formed at this pH (6.25 ± 0.09) and detention time (30 to 60 min).

There was no clear pattern in the alkalinity data (Table 17) and in most cases there was no significant change in this parameter through the units. Again there was a difference in raw water alkalinity between the high water flowrate and the low water flowrate runs (41 ± 3.9 mg/L as a $CaCO_3$ vs. 79 ± 4.0 mg/L as $CaCO_3$, respectively). The only decrease in alkalinity occurred at the low water flowrate and an A:W ratio of 20:1 (81 ± 0.64 to 71 ± 1.0 mg/L as $CaCO_3$), but is likely the result of variations in the analytical technique.

The pHs of the high and low water flowrates were 6.3 ± 0.09 and 6.5 ± 0.20, respectively. In all cases, except the high water flowrate and A:W ratio of 2:1, there was a significant increase in pH through the units at α = 0.05 (except low water flowrate and A:W ratio of 3:1 which was at α = 0.10) and at α = 0.01 (except high water flowrate and A:W ratios of 3:1 and 10.5:1). As shown in Table 18, the effluent pH increased as A:W ratio increased. The increases probably resulted from an increased removal of CO_2 from the water with larger air flowrates.

ECONOMICS

The cost estimates for the diffused bubble aeration system used at Derry, NH are shown in Table 19. The economic analysis was performed using the techniques described in Section 4. The total production cost for the diffused bubble system used at Derry, treating 37.85 m^3/min, was estimated to be $0.64/1,000 gallons. If pretreatment for iron and manganese via ion exchange is required (see Section 4) then the total production cost with pretreatment would be $2.14/1,000 gallons. The capital and O&M costs for the diffused bubble system are in close agreement with those reported by Malcolm Pirnie, Inc. (1988). The latter study estimated the capital and O&M costs for a plant treating an average daily flow of 49 m^3/day (227 m^3/day design flow) to be $12,200 and $1,000/year, respectively.

The major costs of the diffused bubble system result from the capital costs of process equipment and the resulting annual O&M costs of this equipment. The diffused bubble performance data obtained in this study indicates that the same process equipment can achieve comparable radon removals at flows up to 150 m^3/day (0.10 m^3 H_2O/min) which is far in excess of the 38 m^3/day design flow. Therefore, the production costs for diffused

TABLE 16. INFLUENT AND EFFLUENT TURBIDITY FOR THE HIGH WATER FLOWRATE DIFFUSED BUBBLE SYSTEM RUNS (A:W RATIO 7:1 AND GREATER)

A:W Ratio	Turbidity (NTU) Influent	Turbidity (NTU) Effluent	Statistical[*] Relationship
7:1	0.13 ± 0.04	1.21 ± 0.25	Inf < Eff α^{+} = 0.05 & 0.01
10.5:1	0.18 ± 0.05	1.29 ± 0.12	Inf < Eff α = 0.05 & 0.01
15:1	0.27 ± 0.01	1.33 ± 0.06	Inf < Eff α = 0.05 & 0.01
20:1	0.36 ± 0.01	2.23 ± 0.81	Inf < Eff α = 0.05 & 0.01

[*] As assessed by Student's t test.

[+] α: significance level.

TABLE 17. STATISTICAL COMPARISON* OF INFLUENT AND EFFLUENT ALKALINITY IN THE DIFFUSED BUBBLE SYSTEM

Water Flowrate	A:W Ratio	Statistical Relationship	Significance Level α
Low	2:1, 3:1, 5:1, 7:1, 10.5:1, 15:1	Inf = Eff	0.05 & 0.10
	20:1	Inf > Eff	0.05 & 0.01
High	2:1, 3:1, 5:1, 7:1	Inf = Eff	0.05 & 0.10
	10.5:1	Inf = Eff Inf < Eff	0.05 0.10
	15:1	Inf > Eff Inf = Eff	0.05 0.01
	20:1	Inf = Eff	0.05 & 0.10

*As assessed by Student's t test.

TABLE 18. EFFLUENT pHs FOR DIFFUSED BUBBLE RUNS

Water Flowrate	A:W Ratio	Effluent pH
Low	2:1	6.8 ± 0.02
	3:1	6.8 ± 0.11
	5:1	7.5 ± 1.08
	7:1	7.4 ± 0.05
	10.5:1	7.7 ± 0.04
	15:1	7.7 ± 0.05
	20:1	7.8 ± 0.09
High	2:1	6.5 ± 0.04
	3:1	6.8 ± 0.18
	5:1	6.9 ± 0.07
	7:1	6.9 ± 0.16
	10.5:1	6.8 ± 0.15
	15:1	7.4 ± 0.13
	20:1	7.2 ± 0.20

TABLE 19. COST ESTIMATE FOR THE DIFFUSED BUBBLE SYSTEM AT DERRY, NH

Item	Total Cost (2nd Quarter 1989 dollars)*
CAPITAL COSTS	
DIRECT COSTS	
Process Equipment	
3 - Polyethylene tanks ($370.94/tank)	$ 1,113
1 - Fuji ring compressor	606
1 - Float valve	155
1 - Control panel	207
1 - Air pump filter assembly	31
1 - Air vent assembly	108
1 - Air manifold	205
3 - Air diffuser assemblies ($358.67/diffuser)	1,076
1 - Booster/transfer pump+	800
1 - Rockwell flow rate recorder/totalizer	1,036
Total Process Equipment Cost	$ 5,337
Support Equipment	
Installation	150
Plumbing	1,123
Electrical	700
Total Support Equipment Cost	$ 1,937
TOTAL DIRECT COST	$ 7,310
INDIRECT COSTS	
Sitework	$ 1,097
Engineering	1,097
Contractor Overhead and Profit	877
Legal and Financial Fees	183
Interest	439
Contingencies	$ 1,097
TOTAL INDIRECT COST	$ 4,790
TOTAL CAPITAL COST	$12,100
AMORTIZED CAPITAL COST (Annual)	$ 1,326

(continued)

TABLE 19 (continued)

Item	Total Cost (2nd Quarter 1989 dollars)[*]
ANNUAL O&M COSTS	
Power	310[**]
Maintenance	
Mechanical	391
Non-mechanical	57
Labor	110
Administrative	134
TOTAL ANNUAL O&M COST	$ 1,002
TOTAL ANNUAL COST	$ 2,328/year
TOTAL PRODUCTION COST (Design flow 37.85 m^3/d)	$0.64/1,000 gallons
PRETREATMENT COST	$1.50/1,000 gallons
TOTAL PRODUCTION COST WITH PRETREATMENT	$2.14/1,000 gallons

[*] Based on ENR Construction Cost Index, Base Year 1967 = 100.

[+] Although a transfer pump was not required at this site, it is likely to be required at most locations.

[**] Cost based on input power requirements of 1 hp each for the air blower and the transfer pump, operating period of 5.6 hr/day for each, and an electric rate of $0.10157/kW-hr.

bubble systems may decrease markedly as design flows increase. (N.B., the most cost effective blower is a 0.38 kW constant speed blower for the range of flows evaluated in this study, so capital and power costs for the blower would be constant.)

A major assumption implicitly made in the economic analysis is that the radon present in the off-gas will disperse in the wind currents and will not require additional treatment. Although the economic analysis does account for the cost of venting the radon above the average home roof line (i.e., 11 m), no additional air treatment costs are included. If subsequent treatment is required to meet federal or state air quality standards, then the cost estimates could be significantly higher.

SECTION 6

PACKED TOWER

INTRODUCTION

Packed tower aeration has several advantages when used as a treatment technique for removing radon from water. It is rapid, requiring contact times of seconds to minutes. It also uses fairly low volumetric A:W ratios, often less than those used for diffused bubble aeration. In addition, a tower can be easily retrofit to most buildings without requiring much floor space. Unlike GAC systems, it does not concentrate radon and its progeny and as a result is not a source of gamma emissions. The disadvantages most often associated with packed tower aeration include: high costs of operation (aeration system), release of radon to the atmosphere, potential oxidation of iron, accumulation on and sloughing of bacteria from the packing media, and the need for additional pumping or repressurization of water. Other potential problems may be the excessive noise of the air blower and in northern climates, freezing conditions causing ice formation with the tower and pipes.

Cummins (1987) evaluated the radon removal efficiency of packed tower aeration for small community water supplies using several packing heights (0.15 to 5.33 m), and A:W ratios (0.05:1 to 15:1). These field studies used a 0.61 m diameter tower packed with plastic saddles receiving an influent radon activity ranging from 4,100 to 6,200 pCi/L. At A:W ratios of 2:1, 5:1 and 15:1, the tower achieved 95, 99.1 and 99.4 percent removal, demonstrating it could efficiently remove radon yielding effluent activities <50 pCi/L. Dixon and Lee (1987, 1989) reported radon removals of 95-97% using a packed tower 4.88 m in height and 1.37 m in diameter with an A:W ratio of 50:1 (packing medium unknown). The influent radon activity to the tower, which was designed to remove volatile organic compounds, was 646-783 pCi/L.

METHODS AND MATERIALS

The research was conducted in two phases. Phase I evaluated the effect of various parameters on the tower's performance including liquid loading rate, volumetric A:W ratio, packing media and influent radon activity. Phase II involved continuous operation over 6 to 8 hr periods at one condition.

Description of Packed Tower System

The packed tower aeration system, designed by North East Environmental Products (Lebanon, NH), treated the water supply at the same mobile home park in Mont Vernon, NH used in the GAC study. The system consisted of an 5.49 m tall, 0.30 m diameter, stainless steel tower containing randomly packed plastic media (Figure 81). It was supported within a wooden housing and set on a concrete base. Water was provided to the top of the tower directly from 1 or 2 bedrock wells via 3.18 cm PVC piping. A nozzle located 15.24 cm above the top of the media distributed the water. Air entered the tower 1.07 m from the bottom (0.15 m below the media) and was supplied from a manually-triggered American Fan (Fairfield, OH) single phase industrial electric air blower via a 10.16 cm plastic tube connection. The countercurrent air flow aided in the release of the radon from the water. A 10.16 cm plastic hose connected to the top of the tower carried the effluent gas (off-gas) outside. A 0.63 cm plastic tube was inserted 15.24 cm into the end of the hose and was used to sample radon activity in the off-gas.

The treated water was collected in a reservoir at the base of the tower below the media. Electrodes were placed in this reservoir so that when the water level reached a height of 0.91 m, a transfer pump would empty it to a level of 22.86 cm. The treated water was pumped into the community's 7,571 L atmospheric holding tank. During Phase II, the effluent water was wasted to a field behind the pumphouse. Wasting was necessary to operate the system continuously without flooding the pumphouse.

An in-line water flowmeter was located in the 3.18 cm PVC pipe that carried the effluent water to the atmospheric tank or outside the pumphouse. A rotometer capable of measuring water flowrates ranging from 0.97 - 9.65 m^3/hr was located on the influent line to the tower. Pressure and temperature gauges were placed in the influent and effluent water lines and in the influent air flow line. A pitot tube was placed in the influent air flow line and attached to a pressure gauge to measure air flowrates, which were regulated by an in-line globe valve.

The tower contained 3 sampling ports located within the media, as well as influent and effluent sampling ports. Each sampling port consisted of a stainless steel, half cylinder trough (0.95 cm diameter) which protruded about 15.24 cm into the tower. Media sampling ports were located 0.46 m, 1.98 m, and 3.81 m below the influent nozzle. Plastic tubing (1.27 cm diameter) carried the sample water from each port to a sampling board located at the base of the tower. The influent port was tapped into the influent 3.81 cm PVC line just before the water entered the tower and the effluent port was tapped into the reservoir 5.08 cm from the bottom. 0.97 cm plastic tubing transported the influent and effluent water from the ports to the sampling board. A 10.16 cm diameter plastic funnel was attached to the free end of each sampling hose. During sampling, the mouth of the funnel was upright so that the water from the hose filled it and overflowed its lip. This facilitated collection of radon samples even at low water flowrates.

Experimental Design

The Mont Vernon raw water had an average radon activity of 190,134 ± 48,237 pCi/L throughout the packed tower study. Phase I of the study consisted of several separate 3 hr runs designed to determine the tower's radon removal efficiency for a variety of operating conditions. A second phase tested one of these conditions over two 8 hr and one 6 hr continuous

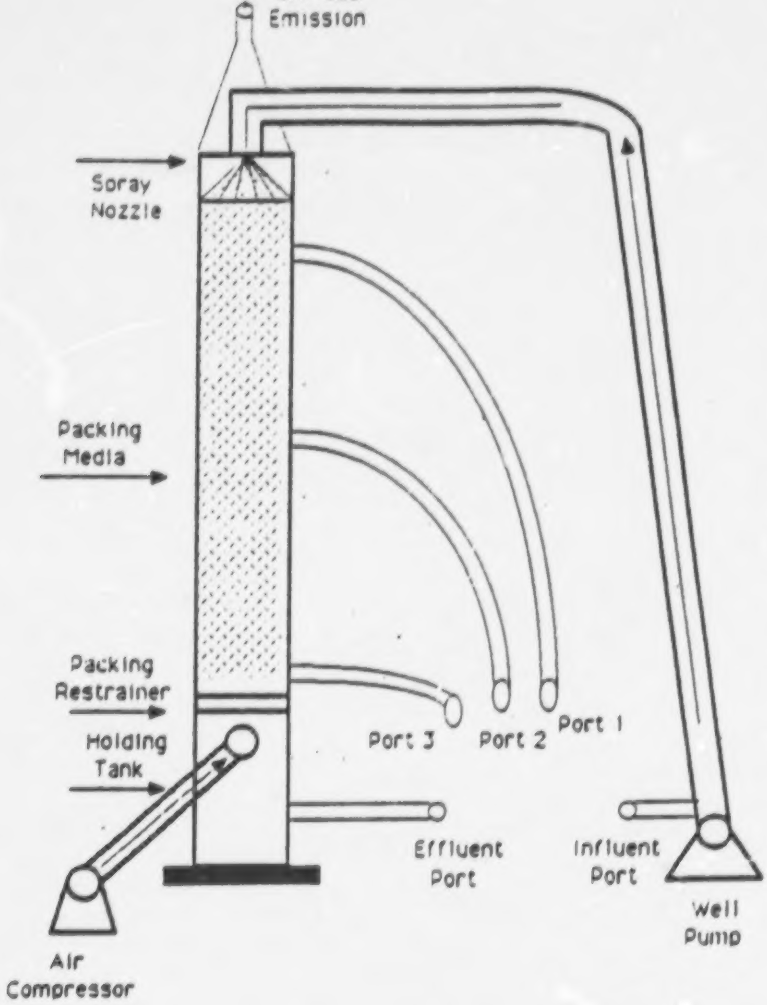

Off-Gas
Emission
Spray
Nozzle
Packing
Media
Packing
Restrainer
Holding
Tank
Port 3
Port 2
Port 1
Effluent
Port
Influent
Port
Well
Pump
Air
Compressor

Figure 81. Schematic of the packed tower aeration system. Mont Vernon, NH.

flow periods. The duration of these continuous flow periods was based on access to the water supply as regulated by the community.

Comparisons were made among combinations of four parameters: packing type, packing height, liquid loading rate, and volumetric A:W flow ratios. During Phase I, three types of packing media were tested: 5.08 cm diameter Glitsch^R mini rings (packing factor (C_f) = 30), 2.54 cm diameter Glitsch^R saddles (C_f = 33), and 2.54 cm diameter Koch^R pall rings (C_f = 45) using the ports installed at the 3 depths within the tower to monitor removal. The packing heights were 0.30, 1.83, 3.66 and 3.75 m when the tower was filled with mini rings and saddles, and 0.15, 1.68, 3.51, and 3.60 m when filled with pall rings. Two liquid loading rate ranges were obtained by adjusting the liquid flow into the tower. Manually activating one well provided the low flowrate range of 0.18 to 1.07 m^3 H_2O/hr (liquid loading rate of 2.5 to 14.7 $m^3 H_2O/m^2$-hr), while 2 wells provided a high flowrate range of 0.74 - 2.55 m^3 H_2O/hr (liquid loading rate of 10.2 to 35.0 m^3 H_2O/m^2-hr). Variations in flowrate were the result of the low yield of the wells at Mont Vernon during this testing period. Air flows were adjusted to obtain approximate volumetric A:W ratios of 20:1, 10:1, and 5:1 for each liquid flowrate range, media type, and packing height. A:W ratios of 2:1 and 1:1 were subsequently tested for the high flowrate, using the pall rings.

Phase II was conducted using both wells with an average water flowrate of 1.46 ± 0.20 m^3 H_2O/hr and an A:W ratio of 2:1. Pall rings were used as packing media since they were the last medium tested in Phase I and had yielded good radon removals. The influent radon activity during this phase of the research averaged 203,675 ± 22,412 pCi/L.

Sampling Plan

The parameters monitored for the packed tower aeration system are shown in Table 20. During Phase I, water samples/measurements for radon, alkalinity, turbidity, iron, manganese, temperature, pressure, and flowrate, and air samples/measurements for pressure, temperature, and flowrate were taken at 15, 30, 60, 120, and 180 min. Air temperature, barometric pressure, and relative humidity were measured inside the pumphouse at the beginning of each run. Background air samples for radon analysis were taken at the beginning of each run, and off-gas emissions were monitored once each sampling run, usually after two hours of operation.

During Phase II, water samples/measurements for radon, alkalinity, turbidity, iron, manganese, temperature, pressure, and flowrate and air samples/measurements for pressure, temperature, and flowrate were all taken at initial startup and each hour thereafter (over the 6 to 8 hr run time). These parameters were monitored during Phase II to determine the effects of aeration over longer operational periods. Relative humidity, temperature, and barometric pressure of the air in the building were measured at the beginning of each run. Background air samples for radon were taken at the beginning of each run and off-gas emissions were measured halfway through and at the end of each continuous operation period. All temperatures were measured using

TABLE 20. PACKED TOWER AERATION SAMPLING PARAMETERS

	Radon	Alkalinity	Turbidity	Iron/Mang.*	Flowrate	Temp.+	Pressure	Barometric Pressure	Relative Humidity
Water Samples									
- Influent	X	X	X	X		X	X		
- Port 1	X								
- Port 2	X								
- Port 3	X								
- Effluent	X	X	X	X	X	X	X		
Air Samples									
- Influent					X	X	X		
- Building						X		X	X
- Effluent	X								

---

*Iron and manganese.

+Temperature.

in-line thermometers. Water and air pressures were also measured using in-line gauges. Water flowrates were measured using an in-line flowmeter. Air flowrates were measured using a pitot tube placed in the influent air line, which was attached to a Dwyer^{R} magnehelic differential pressure gauge (Michigan City, IN). Pitot tubes were placed in PVC pipes of sizes 1.27, 2.54, 5.08 and 10.16 cm, depending on the required air flow. The magnehelic had a gauge reading 0 to 1.27 cm (water).

RESULTS AND DISCUSSION

Radon

Phase I - Evaluation of A:W Ratio, Packing Media and Water Flowrate--

Overall System Performance--The experimental design for the packed tower experiments was supposed to evaluate the effect of water flowrate (liquid loading), air flowrate (A:W ratio) and packing media on radon removal. Unfortunately, during Phase I, the wells at Mont Vernon were pumping erratically (summer and early fall 1988). At that time, New Hampshire had received little rainfall and the groundwater table was depressed. In spite of the fact that the wells in Mont Vernon were deep (61-91 m) and believed to be in a bedrock aquifer, they appeared to be affected by the lower water table. Their yields were low and the radon activities associated with the water fluctuated greatly on a daily basis.

As a result, the liquid loading rates (i.e., water flowrates) and radon activities for packed tower runs made during Phase I were not comparable (Figures 82 and 83). For example, the high water flowrate runs had significantly different flowrates for each packing medium within a given A:W ratio. Similarly, the runs with a given packing medium at the different A:W ratios often had very different radon activities in the raw water further confounding comparisons.

The only factor which was well regulated among the trials was the air flowrate which could be controlled by the sampling team. In addition, influent radon activities were often similar for more than one of the sampling times within a run at a given test condition (Figure 84). For purposes of data analysis when there was <8.0% difference between the raw water radon activities in a given run they were considered to be similar and were pooled for use in statistical analysis.

Determinations of treatment efficiency could not be made based on the percent mass of radon removed per time [i.e., $(Q_w \times C_{inf} - Q_w \times C_{eff})/Q_w C_{inf} \times 100$] because of the constant variations in water flowrate. Therefore, all percent removals were expressed in terms of the change in radon activity [$(C_{inf} - C_{eff})/C_{inf} \times 100$]. Due to the very short residence time of water in the tower, comparison of influent and effluent activities at a given sampling time was justified because short term variations in influent activity were minimal.

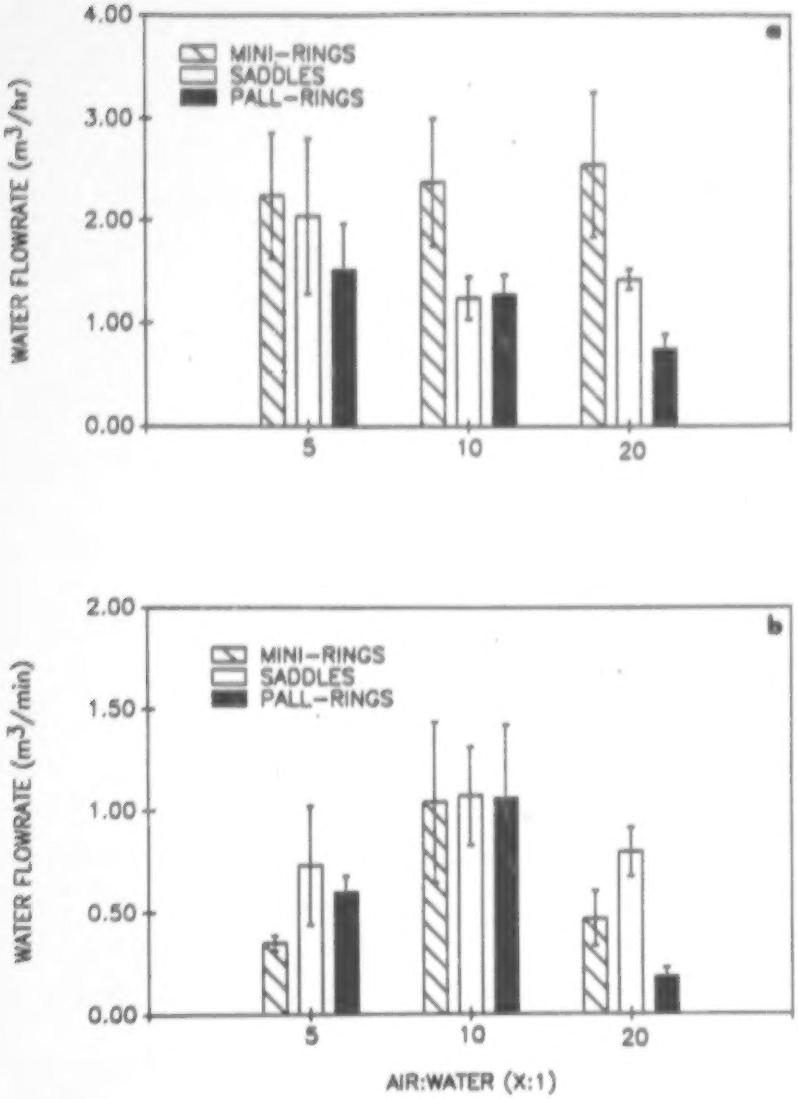


Figure 82. Phase I - Packed tower aeration. Mont Vernon, NH. Water flowrate as a function of A:W ratio for each packing medium. (a) high water flowrate, (b) low water flowrate. Note differences in scale on ordinate.

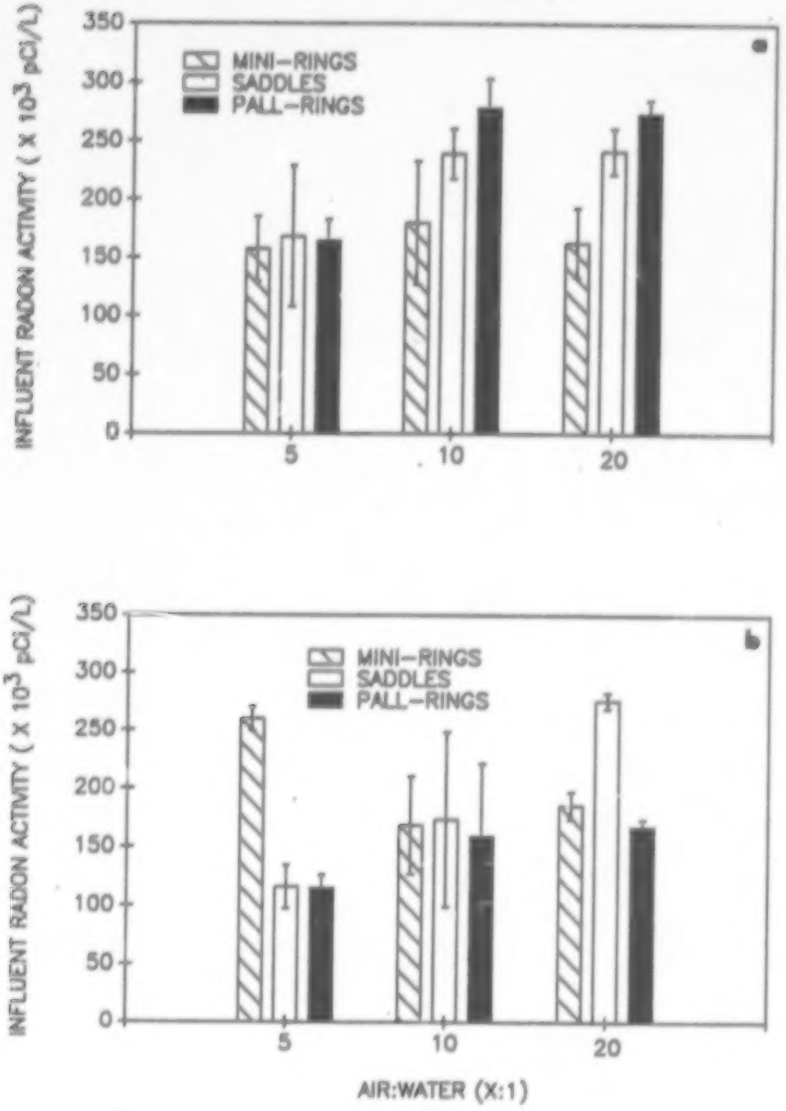


Figure 83. Phase I - Packed tower aeration. Mont Vernon, NH. Influent radon activity as a function of A:W ratio for each packing medium. (a) high water flowrate, (b) low water flowrate.

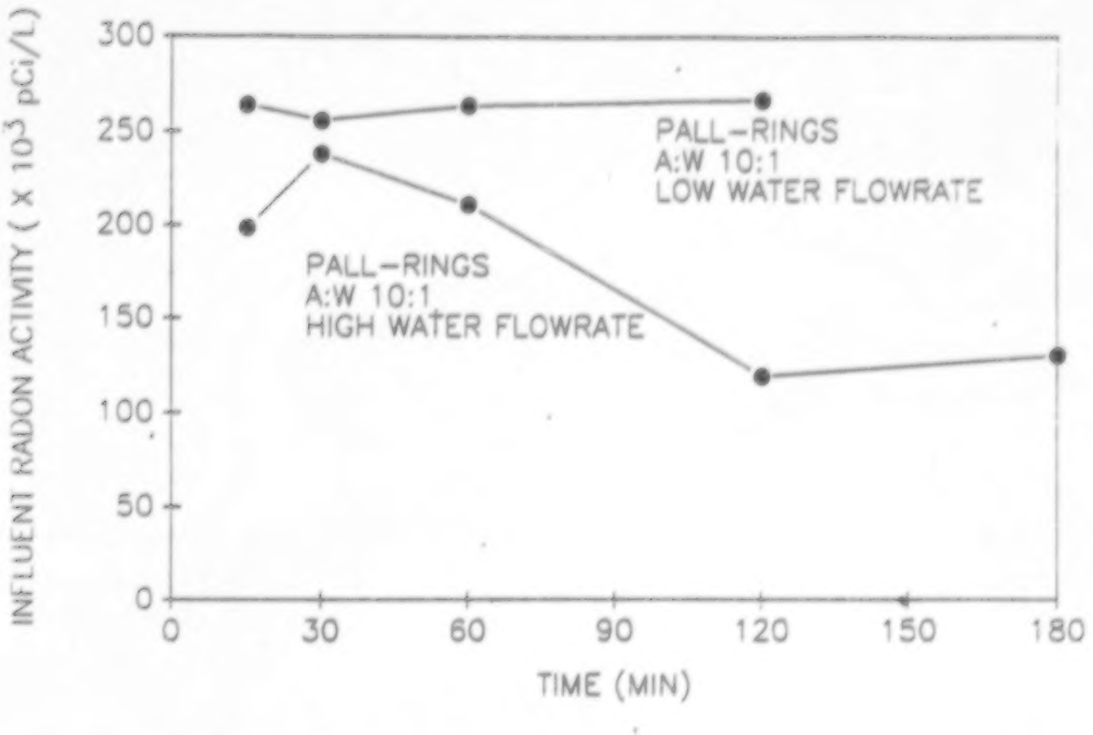


Figure 84. Phase I - Packed tower aeration. Mont Vernon, NH. Influent radon activity as a function of time for two runs.

Though the fluctuations in water flowrate and radon activity prevented comparisons of the effects of A:W ratio and packing type, there was relatively little difference in the overall percent radon removal observed among the conditions tested (Figure 85). For a packed tower aeration system, mass transfer is usually less sensitive to changes in air flowrate than to changes in liquid loading rate and interfacial surface area provided by the media (Kavanaugh and Trussell, 1980). The percent radon removals only varied from 92.7 to 99.8, which is surprising considering the variation in water flowrate (0.18 to 2.6 m^3/hr), radon activity (115,255 to 278,488 pCi/L) and packing type. The resilience of the packed tower system is encouraging considering that many small communities required to install radon treatment systems may experience variations in water flowrate and radon activity similar to those observed at the Mont Vernon site. At the air flowrates tested, the changes in liquid loading and mass loading of radon probably did not impact the effluent activity because (i) radon is a highly volatile gas and thus is readily transferred from water to air in the tower and (ii) the tower (~3.66 m of packing media) was deep enough to compensate for the large variations in loading (i.e., the tower contained far more theoretical transfer units than required since radon is so volatile).

It is important to note that the height of the packed tower (5.49 m) and the range of liquid loading rates (2.5 to 35 m^3/m^2-hr) used in this study were in the lower range of those used in water treatment for volatile organic contaminant removals where typical heights and liquid loadings are 4.3 to 12.2 m and 13.7 to 105 m^3/m^2-hr, respectively. Therefore, the effects of using greater tower heights and/or liquid loadings on effluent radon activity should be considered, particularly for systems required to meet stringent effluent radon design goals.

Performance within Tower Sections--Figure 86, profiles of percent radon removal obtained within the tower during the various runs, shows that a significant amount of the treatment occurred in the top 0.30 m of the mini rings (59.1 - 72.5%) and saddles (45.4 - 53%). In this section, the driving force (C_L - C_G) for mass transfer of radon from the liquid concentration (C_L) to the gas phase concentration (C_G) was the greatest. In addition, the end effect of the nozzle spraying the water over the packing probably aided in release of the radon to the air. Likewise, the water flowing through the very top section of the packing was experiencing the most turbulent flow conditions in the tower. It is well documented in the literature (Perry and Chilton, 1983) that end effects in packed tower air strippers resulting from liquid distributors, free fall and turbulence can increase removal of the contaminant. This pattern is less clear with the pall ring (38.9 - 47.3%) data in which the water only passed through 0.15 m of packing before reaching the first sampling port.

Through the next approximately 1.52 m of mini-rings and saddles, the radon removal efficiency decreased, ranging from 23.2 to 42.0%. With the pall rings, the removal efficiency of the middle section was as good or better than the top section (43.0 - 50.6%). It is possible that most of the removal still occurred in the top 0.30 m of the pall rings, but because of differences in packing heights and sampling depths, this could not be determined. The last

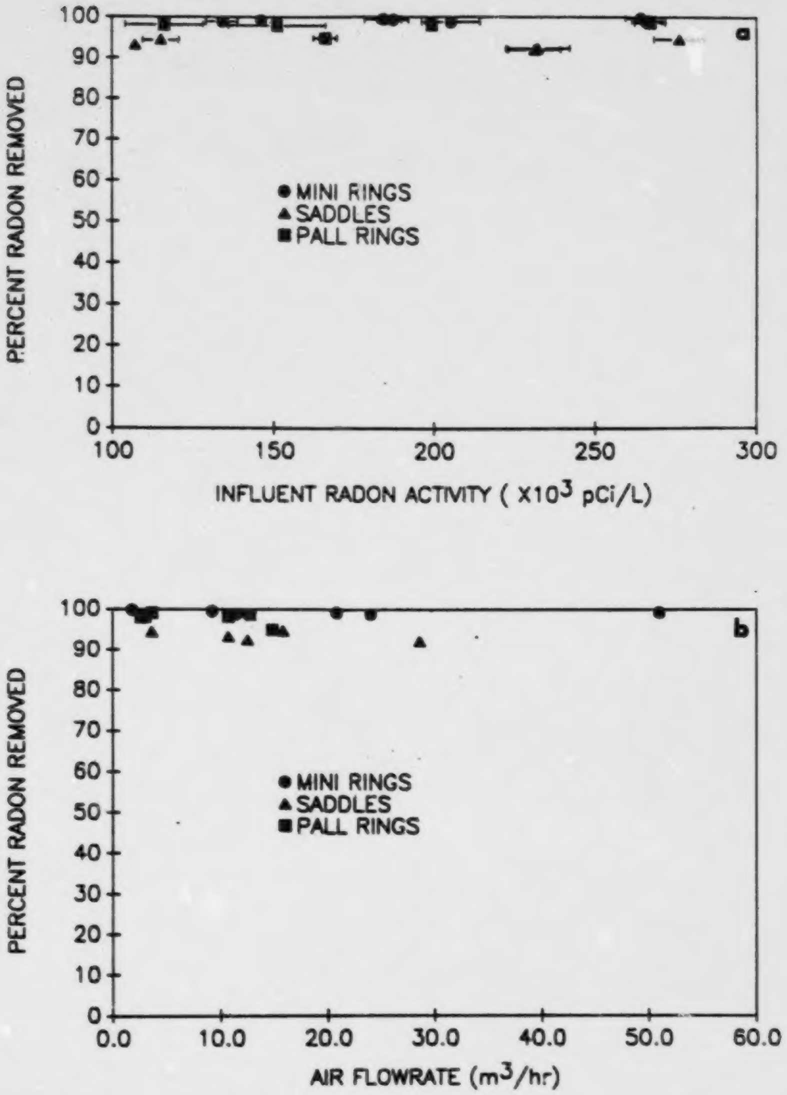


Figure 85. Phase I - Packed tower aeration. Mont Vernon, NH. Percent radon removal as a function of influent radon activity (a) and air flowrate (b) for each condition.

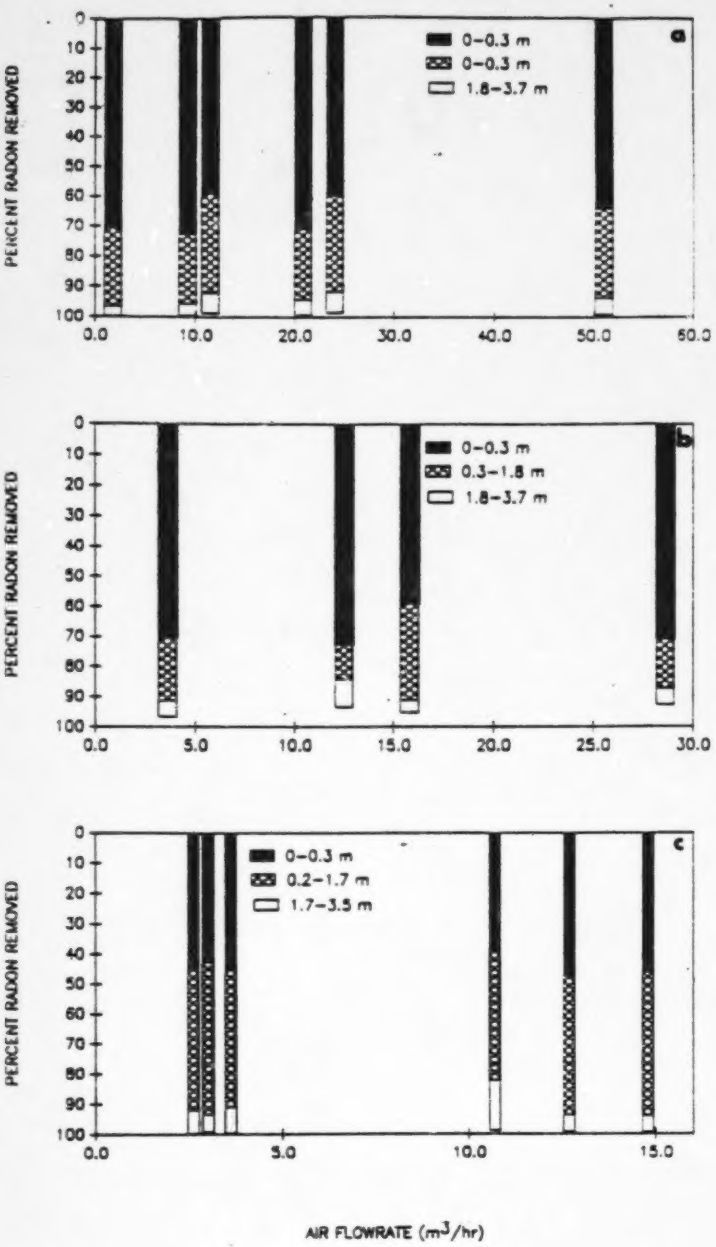


Figure 86. Phase I - Packed tower aeration. Mont Vernon, NH. Percent radon removal as a function of air flowrate within the tower for mini rings (a), saddles (b) and pall rings (c).

1.83 m of packing, in all cases, removed less than 10% of the radon from the water, except with the pall rings at an air flowrate of 10.7 m³/hr (16.9%). The lower radon removal efficiency with depth in the tower probably resulted from the decreased mass transfer due to the lower driving force ($C_L - C_G$). In addition, the liquid flow conditions in these sections were probably far less turbulent than in the top section, especially because there was no redistribution of the liquid flow within the tower at mid-depth. The decreased turbulence can increase the liquid film thickness, which in turn decreases the mass transfer of radon from the water to the air.

Mass Transfer Coefficients--The global liquid mass transfer coefficient, K_La, within a packed tower can be expressed as a function of (i) the molecular diffusion coefficient of the contaminant in water (D_w), (ii) the viscosity (μ_w) and density (ρ_w) of the water, (iii) the liquid loading rate (L), and (iv) empirical constants (m and n) dependent on the type and size of the packing media as shown by the correlation developed by Sherwood and Holloway (1940):

$$K_La = [m(\frac{L}{\mu_w})^{(1-n)} (\frac{\mu_L}{\rho_w D_w})^{1/2}] D_w \qquad \text{(eq. 8)}$$

K_La is also dependent on the water temperature which affects μ_w, ρ_w and D_w. Therefore, the K_La within a packed tower operating at a given liquid loading rate with a specific randomly distributed packing is not a function of packing height. However, the K_La for a given system can be calculated based on data collected from continuous flow operation by solving the mass balance-derived packed tower equations:

$$R = HG/P_tL \qquad \text{(eq. 9)}$$

$$NTU = R/(R - 1)\ln[(C_{in}/C_{out}(R - 1) + 1)/R] \qquad \text{(eq. 10)}$$

$$HTU = Z/(NTU) \qquad \text{(eq. 11)}$$

$$K_La = L/(HTU)C_m \qquad \text{(eq. 12)}$$

where Z = packing height, NTU = number of transfer units, R = stripping factor, H = Henry's Constant, P_t = ambient pressure, HTU = height of a transfer unit, L = liquid loading rate, G = air loading rate, K_La = overall mass transfer coefficient, and C_m = density of the liquid. (N.B., the units for Henry's constant must be in atmospheres in this equation).

The K_La values calculated for the top sections of the tower with all of the media (Figure 87) were substantially higher than those for the lower two

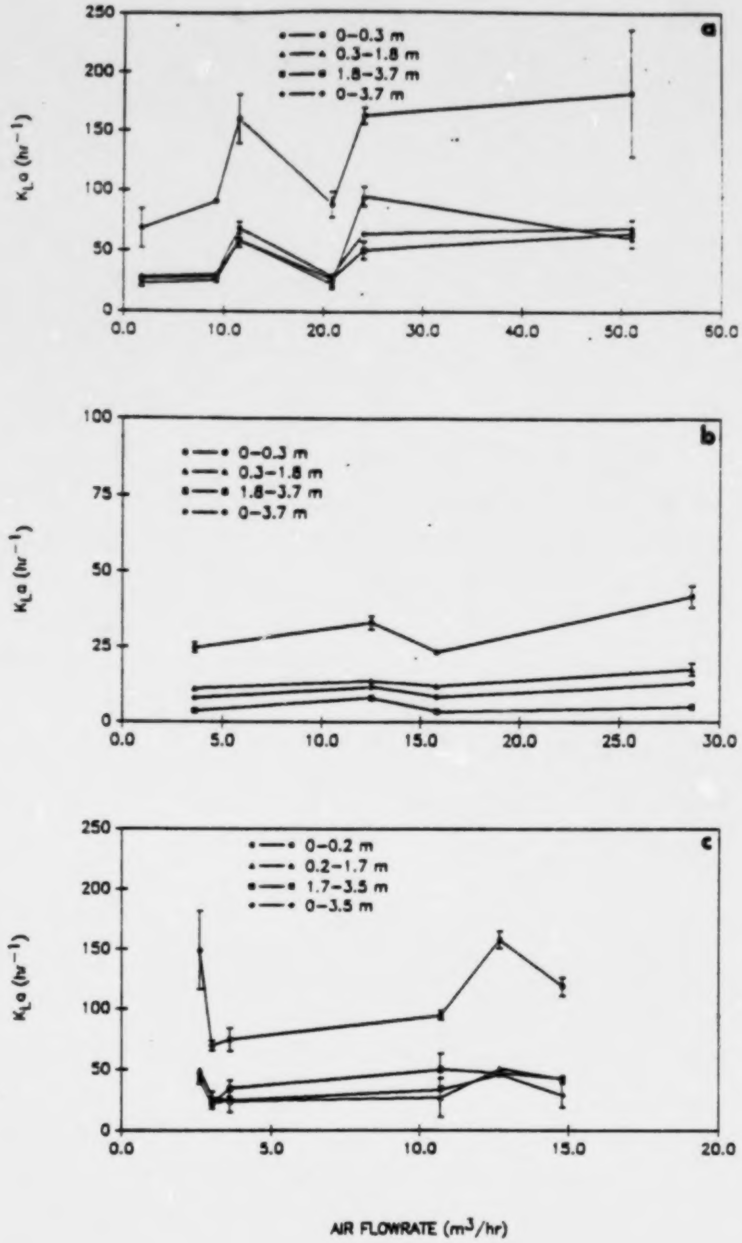


Figure 87. Phase I - Packed tower aeration. Mont Vernon, NH. K_La as a function of air flowrate within the tower for mini rings (a), saddles (b) and pall rings (c). Note differences in scale on ordinate and abscissa.

sections and the overall K_La. (The calculated K_La values only serve as an indication of the true K_La because of the variations in the liquid loading rate during the runs. The values shown were calculated using the average L for each run.) These higher K_La values result from the end effects of the spray nozzle and the turbulent flow in the top section. As seen in Figure 87, the K_La values for the bottom two sections and the overall K_La were very similar and are a better representation of the mass transfer coefficients for the packed tower operating at a given liquid loading rate and with a specific packing medium. (Statistical comparisons were not justified because of the variations in L).

Factors Affecting Packed Tower Operation--The data indicate that mass transfer limitations may be a major factor in designing towers to achieve very low radon activities. These limitations primarily arise from the change in radon concentration (activity) with depth in the tower reducing the driving force. Therefore, in order to achieve low effluent activities, the packing height (and hence, the contact time) would need to be substantial. This was demonstrated by Cummins (1987), who obtained very low effluent activities (<50 pCi/L) with a raw water activity of 4,100 - 6,200 pCi/L at A:W ratios of 2:1, 5:1 and 20:1, but only with a large packing height (5.33 m). For small communities such as Mont Vernon, which have moderate to high radon activities, packed towers may be impractical because mass transfer limitations will dictate the use of extremely large packing heights to meet a 200 pCi/L MCL. As a result of mass transfer limitations, pilot scale testing over the expected range of water flowrates and if possible, radon activities should be conducted in most cases.

The air intake for the blower used at the Mont Vernon site was located inside the building. This was done to decrease problems associated with pumping very cold outside air into the tower during the winter. However, this practice can have repercussions with respect to radon removal efficiency. In the Mont Vernon pumphouse, the air contained as much as 100 pCi/L of radon. Such a high air activity will decrease the driving force (C_L - C_G) and thus the mass transfer of radon, especially when radon activity in the water is low. Therefore, when designing packed tower systems in northern climates, one must evaluate the cost effectiveness of using cold or preheated outside air versus using pumphouse air which may have a significant radon activity.

Though the variations in water flowrate and influent activity prevented detailed statistical analysis of the performance of the 3 packing media, there was a lower overall percent removal observed during Phase I for the saddles (94.4 ± 1.71%) compared with the mini rings (99.2 ± 0.49%) and pall rings (99.3 ± 0.33%). These differences, also observed in the K_La values for the media, occurred even though the range of water flowrates and raw water radon activities tested were similar (Table 21). When the saddles were removed from the tower, it appeared that water was ponding in this packing. Perhaps the ponding occurred because the saddles are primarily solid pieces of plastic,

TABLE 21. RANGE OF INFLUENT RADON ACTIVITIES AND WATER FLOWRATES DURING PHASE I

Packing Medium	Influent Radon Activity (pCi/L)	Water Flowrate (m^3/hr)
Mini Rings	146,630 - 262,753	0.35 - 2.55
Saddles	114,996 - 276,191	0.72 - 2.04
Pall Rings	116,546 - 267,306	0.18 - 1.52

while the mini and pall rings have many openings to facilitate water and air flow. Further testing is probably warranted to determine if there is a significant difference in radon removal efficiency among the packing media.

Phase II - Radon Removal--

Phase II runs were conducted to determine if the packed tower could operate continuously, achieving similar results to those obtained during Phase I. The packed tower was operated on 3 separate days for 6 to 8 hr during the winter of 1988. The water flowrate was much more constant averaging 1.46 ± 0.20 m³ H_2O/hr. The pall rings were used and the A:W ratio was set at 2:1. The influent activity on the 3 days averaged 181,614 ± 32,781; 202,989 ± 36,429; and 226,422 ± 52,870 pCi/L, respectively. Figure 88 shows overall percent removals of applied radon over time for each of the three days. As observed in Phase I, removal efficiency remained fairly constant regardless of the variation in influent activity. Effluent activities were 4,882 ± 1,659, 3,558 ± 1,311 and 5,173 ± 1,667 pCi/L, respectively. As observed during Phase I with the pall rings, the majority of the radon removal occurred in the top (43.3 ± 2.4%) and middle (46.8 ± 1.3%) sections of the tower (Figure 89), while the bottom 1.83 m removed only a small percentage (7.5 ± 0.41%) of the radon applied. The K_La values were also similar to those obtained at an A:W ratio of 2:1 (pall rings) during Phase I (Figure 90).

The Phase II tests were conducted during the winter, in part, to determine whether colder air and water temperatures would affect performance. During Phase II, air and water temperatures as low as 7°C and 4°C, respectively did not appear to decrease the removal over that obtained during Phase I when the temperatures were 21.2 ± 1.64°C and 13.1 ± 1.4°C, respectively. However, the tower and its piping must be protected from freezing conditions. The tower was enclosed in a wooden structure attached to the Mont Vernon pumphouse. This structure channeled cold ambient air into the building. There were a few extremely cold nights (<-18°C) during the operating period and some of the Schedule 40 PVC piping used to plumb the tower and the glass water flowrate rotometer cracked, even though neither of them contained water. (All water was drained from the system when the tower was not operating.) This experience indicates that in full-scale operation at small communities in northern climates, provisions should insure that pipes and/or towers are insulated to prevent freezing, especially during periods of low or intermittent water flow.

Off Gas Monitoring

Off-gas emissions were monitored to determine the radon activity exiting the tower. In most cases for Phase I and II, the radon activity measured in the air using a Pylon AB-5 monitor was less than the theoretical radon activity (Figures 91 and 92) calculated using a mass balance approach (Q_WC_W/Q_AC_A). There are several explanations for this discrepancy. The wide fluctuations in water flowrate and influent radon activity made calculation of the theoretical radon activity in the off-gas difficult. However, the fluctuations were probably not great enough to account for recoveries consistently less than 50%. There may have been problems associated with the

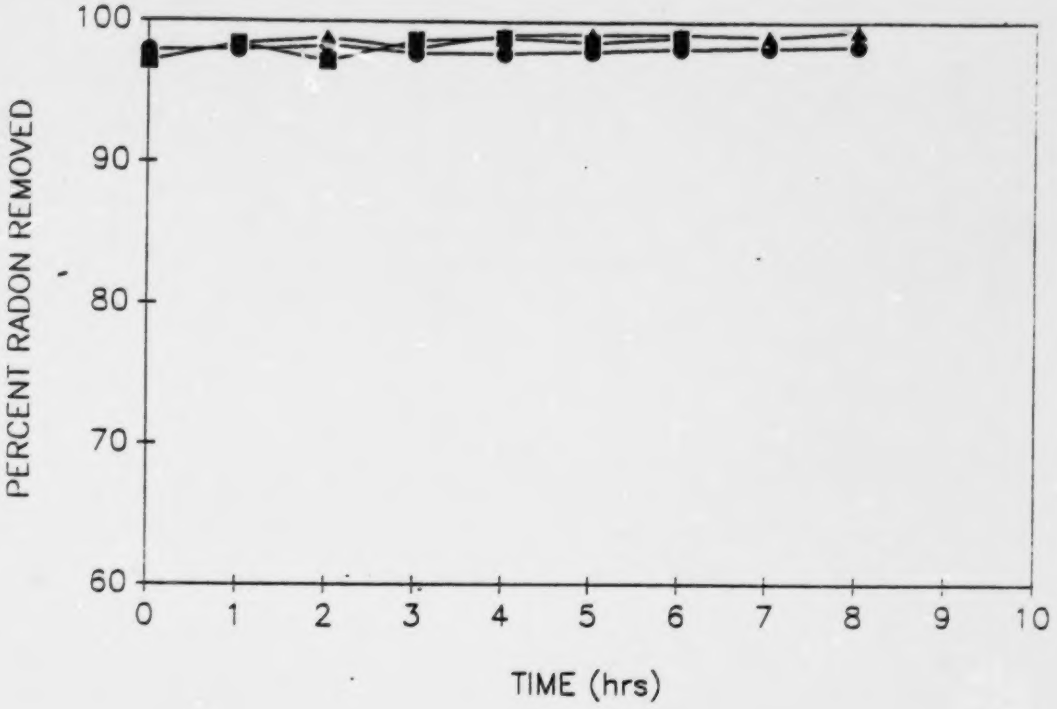


Figure 88. Phase II - Packed tower aeration. Mont Vernon, NH. Percent radon removal as a function of time for each day of testing (pall rings, water flowrate = 1.46 ± 0.2 m^3 H_2O/hr, A:W = 2:1).

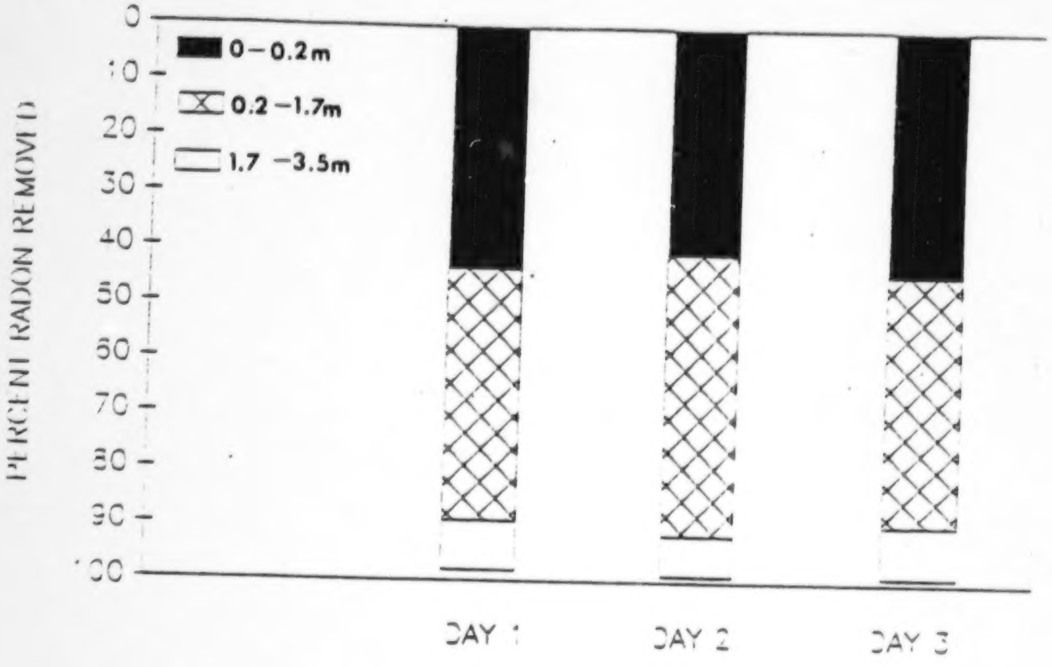


Figure 89. Phase II - Packed tower aeration. Mont Vernon, NH. Percent radon removed within the tower for each day of testing. (Pall rings, water flowrate = 1.46 ± 0.2 m³ H_2O/hr, A:W ratio = 2:1).

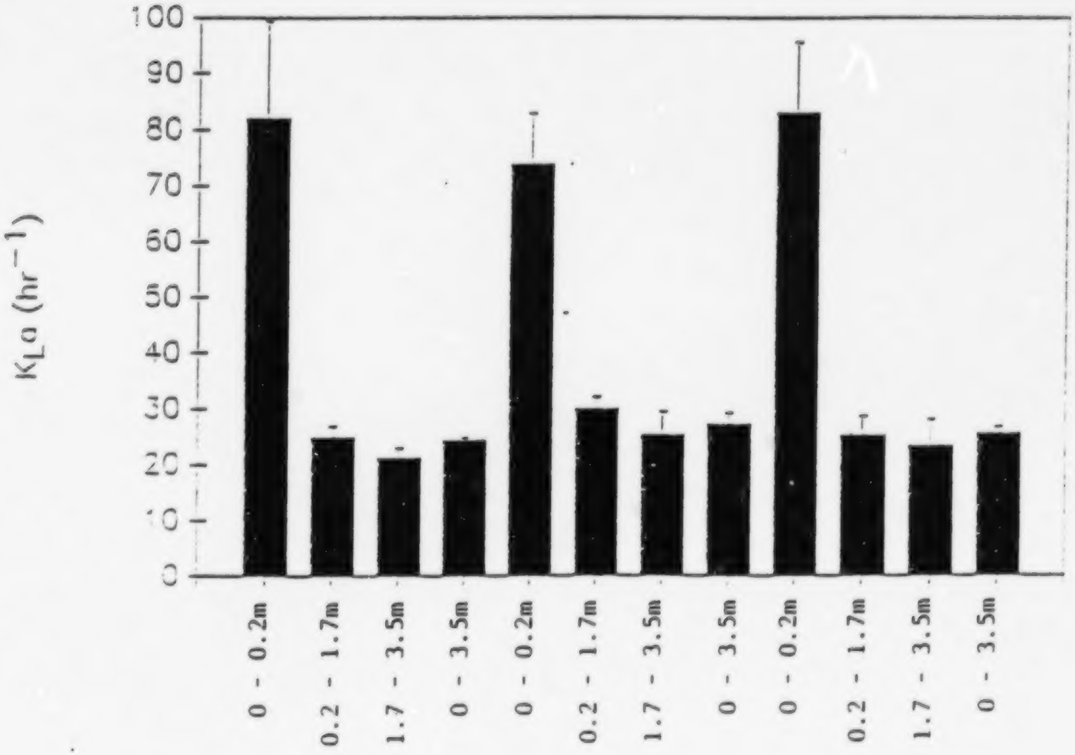
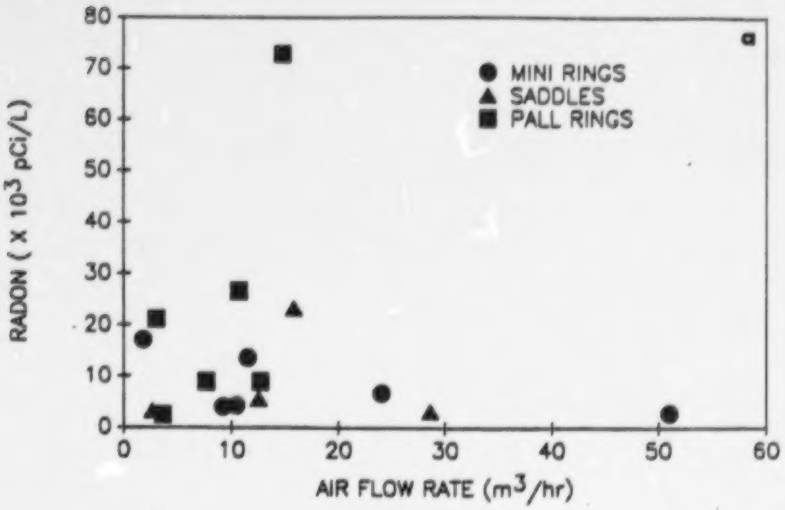


Figure 90. Phase II - Packed tower aeration. Mont Vernon, NH. K_La values within the tower.



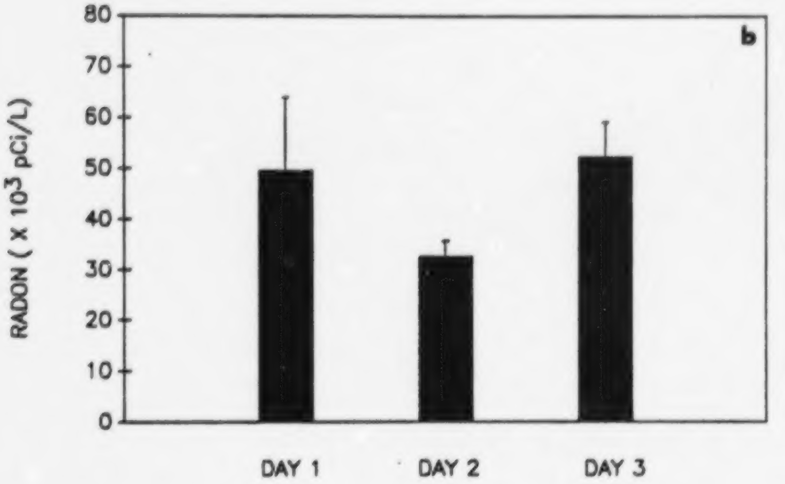


Figure 91. Packed tower aeration - Mont Vernon, NH. Percent off-gas recovery in Phase I as a function of air flovrate for each packing medium (a) and percent off-gas recovery in Phase II (b).

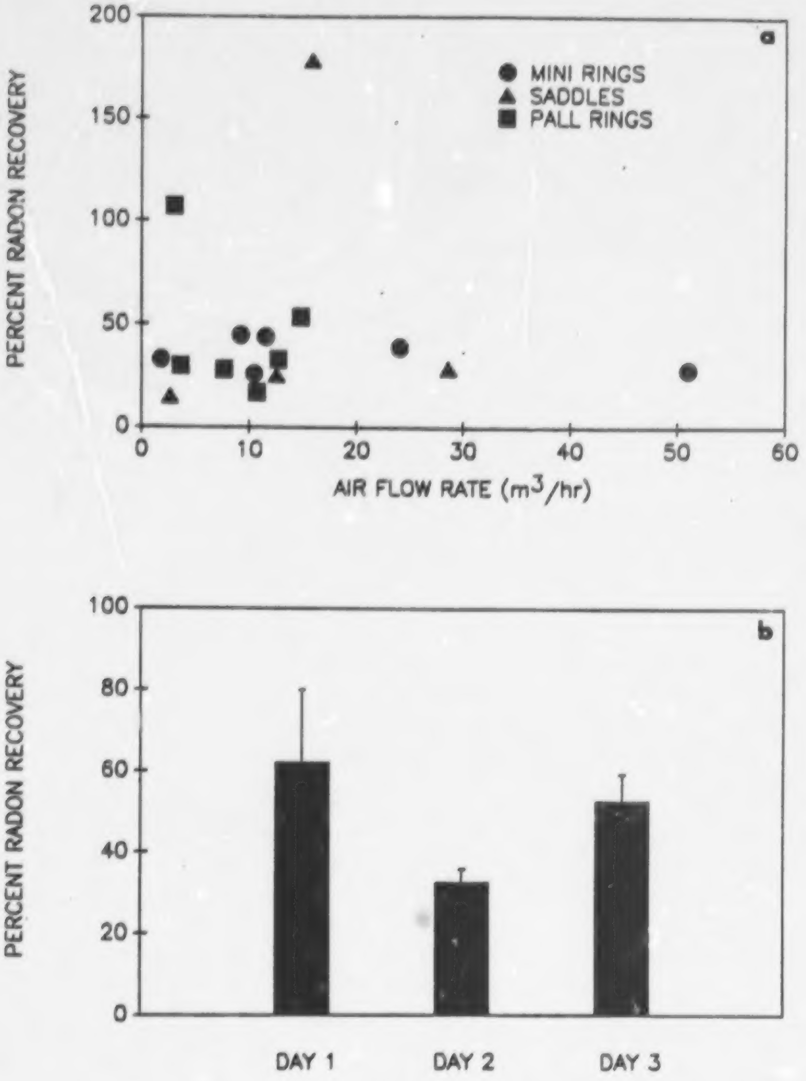


Figure 92. Packed tower aeration - Mont Vernon, NH. Off-gas radon activity in Phase I as a function of air flowrate for each packing medium (a) and off-gas radon activity in Phase II (b).

collection of the air samples. The off-gas exited the top of the tower via a 10.16 cm diameter black plastic hose. The 0.69 cm diameter air sampling tube, attached to the side of the outlet hose, protruded approximately 0.30 m inside it. The inlet of the sampling tube faced into the off-gas flow. It is possible that the boundary layer effects caused the off-gas air to divert around the intake for the Pylon resulting in the low activities recorded.

In all cases, the off-gas radon activities were extremely high (Figures 91 and 92) and would require a large amount of dilution to approach the activity of ambient air (average of 0.1 to 0.15 pCi/L) (NH Bureau of Radiological Health, personal communication, 1986). In future research, the radon activity surrounding the building should be monitored in addition to the emissions at the top of the tower to determine the dilution rate and the effects of the off-gas on air quality.

Iron and Manganese

The influent total iron concentrations ranged from below detection (0.06 mg/L) to 1.70 mg/L. In almost all of the samples taken at Mont Vernon during operation of the packed tower, the influent iron concentrations were below detection. As a result, the effect of the tower on iron oxidation could not be determined. The average total manganese concentrations in the raw water (0.06 ± 0.01 mg/L) did not change significantly through the tower ($\alpha = 0.05$ and 0.01, StT). This is not surprising since the tower's aeration system was not expected to change the total iron and manganese concentrations unless the metals accumulated in the reservoir or within the packing. In future research on the packed tower system, changes in soluble iron and manganese should be determined to see if precipitate formation is occurring. Pretreatment of waters containing significant amounts of iron and manganese may be necessary to prevent accumulation of metal precipitates within the tower system and problems associated with oxidized iron and manganese in the distribution system.

Turbidities, Alkalinities, and pH

Turbidity, alkalinity, and pH were monitored to determine if the packed tower treatment system had any effect on these basic water quality parameters. Influent turbidities ranged from below detection (0.05 NTU) to 0.95 NTU during Phase I and from 0.05 to 2.80 NTU during Phase II. Effluent values ranged from below detection to 0.80 NTU for Phase I and from 0.05 to 2.00 NTU during Phase II, which was not significantly different from the influent ($\alpha = 0.05$ and 0.10, StT). This was expected since the media were not expected to contribute to the turbidity of the water nor were they expected to remove a significant amount of turbidity from the water.

Influent alkalinities to the packed tower ranged from 18 to 32 mg/L as $CaCO_3$ during Phase I and from 20 to 28 mg/L as $CaCO_3$ during Phase II, while effluent alkalinities ranged from 19 to 30 mg/L as $CaCO_3$ during Phase I and from 13 to 24 mg/L as $CaCO_3$ during Phase II. No significant difference was detected between the influent and the effluent alkalinity ($\alpha = 0.05$ and 0.10, StT), except for two runs where the differences were statistically significant at $\alpha = 0.10$, but the change in alkalinity was less than 0.7 mg/L as $CaCO_3$. Changes in alkalinity due to air stripping were not expected at Mont Vernon based on raw water characteristics.

At all conditions, the tower raised the pH of the water significantly ($\alpha = 0.05$ and 0.01, StT). Influent pH values ranged from 6.3 to 6.9 during Phase I and from 6.4 to 6.7 during Phase II, while effluent pH values ranged from 6.6 to 7.7 during Phase I and from 6.9 to 7.5 during Phase II. The aeration process within the packed tower probably caused the increase in pH by removing CO_2 from the water. Even though the pH was raised significantly, the resulting effluent pH was not outside the 6.5 - 8.5 acceptable range for drinking water.

ECONOMICS

Cost estimates for the packed tower treatment system operated in Mont Vernon, NH, were developed using the approach described in Section 6 and are presented in Table 22. These data indicate that the total production cost for the packed tower system was $0.60/1,000 gallons. If pretreatment for iron and manganese is required then production cost estimates would increase to $2.10/1,000 gallons (Section 4). The production cost for the packed tower system is comparable to that reported by Malcolm Pirnie, Inc. (1988). The latter report concluded that the production cost to treat an average flow of 49 m^3 H_2O/day (227 m^3 H_2O/day design flow) would be $0.58/1,000 gallons for 99% radon removal.

It should be noted that a major component of the packed tower system cost at Mont Vernon is related to support equipment costs. Since support equipment includes a variety of site specific factors (e.g., installation), one would expect this cost to be highly variable from site to site. In addition, performance data for the packed tower system indicates that this facility can be operated at flows up to 61 m^3 H_2O/day (i.e., a continuous flowrate of 2.54 m^3 H_2O/hr and still achieve radon removals comparable to those achieved at the 38 m^3 H_2O/day design flow. Therefore, the production cost of the packed tower system may decrease as design flows increase (for the range of flows observed in this study).

In addition, as was discussed for the diffused bubble system (Section 5), the cost estimates for the packed tower aeration system only include venting the radon contaminated off-gas to the air at a height equivalent to the average home roof line (i.e., 10.7 m). Therefore, if treatment of the off-gas for radon is required to meet air standards, system costs could increase significantly.

TABLE 22. COST ESTIMATE FOR THE PACKED TOWER SYSTEM AT MONT VERNON, NH

Item	Total Cost (2nd Quarter 1989 dollars)[*]
CAPITAL COSTS	
DIRECT COSTS	
Process Equipment	
1 - Stainless steel column	$ 511
10 cu. ft. plastic packing media ($\$668/\text{m}^3$)	189
1 - Air flow meter	47
1 - Bete fog nozzle	28
1 - Demister pad	102
1 - Air blower	348
2 - Air pressure gauges ($34.86/gauge)	70
1 - Air vent assembly	100
4 - Temperature gauges ($22.62/gauge)	91
1 - Booster/transfer pump	800
1 - Rockwell flow rate recorder/totalizer	1,036
Total Process Equipment Cost	$ 3,322
Support Equipment	
Installation[+]	2,350
Plumbing	1,133
Electrical	1,381
Total Support Equipment Cost	$ 4,864
TOTAL DIRECT COST	$ 8,186
INDIRECT COSTS	
Sitework	$ 1,228
Engineering	1,228
Contractor Overhead and Profit	982
Legal and Financial Fees	205
Interest	491
Contingencies	$ 1,228
TOTAL INDIRECT COST	$ 5,362
TOTAL CAPITAL COST	$13,548
AMORTIZED CAPITAL COST (Annual)	$ 1,484

(continued)

TABLE 22 (continued)

Item	Total Cost (2nd Quarter 1989 dollars)[*]
ANNUAL O&M COSTS	
Power	232[**]
Maintenance	
Mechanical	252
Non-mechanical	32
Labor	110
Administrative	93
TOTAL ANNUAL O&M COST	$ 719
TOTAL ANNUAL COST	$ 2,203/year
TOTAL PRODUCTION COST (Design flow 37.85 m^3)	$0.60/1,000 gallons
PRETREATMENT COST	$1.50/1,000 gallons
TOTAL PRODUCTION COST WITH PRETREATMENT	$2.10/1,000 gallons

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[*] Based on ENR Construction Cost Index, Base Year 1967 = 100.

[+] Installation includes cost of poured concrete base for tower and cost to remove and reroute existing utilities to facilitate new construction.

[**] Power costs are based on input power of 0.5 Hp for the blower and 1 Hp for the pump, an operating period of 5.6 hr/day for each, and an electric rate of $0.10157/kW-hr.

SECTION 7

LOW TECHNOLOGY ALTERNATIVES

INTRODUCTION

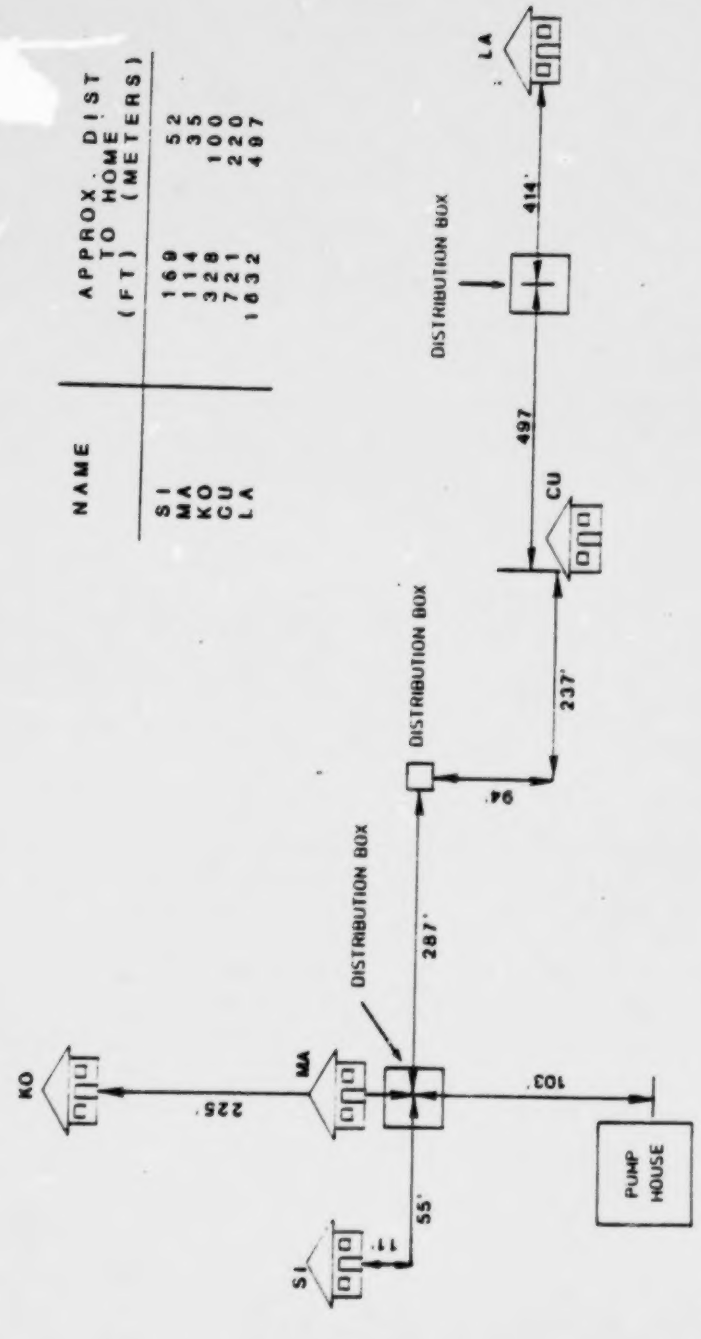
Many very small communities, required to treat their groundwater supplies for radon, will face severe financial problems paying the capital and O&M costs associated with installing GAC, diffused bubble or packed tower aeration systems. The objective of this part of the study was to evaluate the potential of some low technology treatment alternatives to reduce radon levels in drinking water. Low technology systems should (i) be easily retrofit into existing facilities, (ii) require little initial capital investment, (iii) require little routine maintenance, and (iv) have lower operating costs than the conventional radon technologies. In a typical small community water supply in rural New England, water is pumped to an atmospheric storage tank (7571-37,854 L) and then repressurized in a second storage tank (7571-37,854 L). The existing atmospheric storage tank could be readily adapted to low technology radon removal techniques such as coarse bubble aeration. The study included pilot-scale experiments monitoring radon reduction in (i) a distribution system and (ii) in a flow-through reservoir system with a variety of influent control devices and a coarse bubble (>500 µm diameter) aeration system. During this phase of the project, radon was the primary contaminant of concern and thus the techniques evaluated were monitored exclusively for radon removal.

METHODS AND MATERIALS

Loss in a Distribution System

This study was conducted at the Mont Vernon, NH site. The influent radon concentration during this phase of the project ranged between 150,000 to 250,000 pCi/L. The GAC unit was treating the water at Mont Vernon during this period and the GAC effluent ranged between 22,000 to 27,000 pCi/L. The exact design of the distribution system at the site was not available, however, it was determined that the pipes ranged from 3.8 to 5.1 cm diameter (I.D.) and were constructed of black polyethylene. The network (Figure 93) serves approximately 33 mobile homes. Distances between the homes and connections were approximated using a surveyor's tape.

The loss of radon in the distribution system was assessed with both untreated and GAC-treated water at 5 homes (Figure 93). The homes, where samples were collected, were chosen to give an overall assessment of the potential effectiveness of passive removal of radon in the distribution system tested.


KO
225'
MA
DISTRIBUTION BOX
103'
PUMP HOUSE
55'
11'
SI
287'
DISTRIBUTION BOX
DISTRIBUTION BOX
94'
237'
CU
DISTRIBUTION BOX
497
414'
LA

NAME	APPROX. DIST TO HOME (FT)	APPROX. DIST TO HOME (METERS)
SI	169	52
MA	114	35
KO	328	100
CU	721	220
LA	1632	497

Figure 93. Schematic of the Mont Vernon, NH distribution system.

Radon samples were taken in duplicate from the kitchen faucet in each home after all aeration devices were removed. The first two sets of data were collected when the homes were receiving GAC-treated water. When the untreated water test was conducted, the GAC system was by-passed beginning 1.5 to 3 hr before sampling commenced. This lag time insured that the distribution system was completely flushed of treated water. Samples were always taken between 1700 and 1930 hr because of the higher water use during that time and for the convenience of the homeowners. The high use period was chosen because it would simulate the worst case scenario (i.e., shortest detention time in the system).

Loss During Storage

A 1.8 m diameter fiberglass tank was placed outside of the pumphouse at the Derry, NH site. During the study period, a centrifugal pump was used to continuously supply water from the atmospheric storage tank at Derry to the pilot scale fiberglass storage tank located approximately 13.7 m away. In-line brass flow controllers were installed in the pump's effluent line. Three flow controllers were used during the course of the study (1.3 m^3/day, 3.3 m^3/day, and 4.9 m^3/day).

The storage reservoir (Figure 94) was filled to yield a water depth of 0.6 m (total volume = 1628 L as determined in a dye study). The reservoir was covered with a plywood lid which had a hole (0.3 m x 0.30 m) in the center to allow for overhead entry of raw water. A plywood baffle (0.91 m x 0.96 m) was suspended in the water near the effluent port to prevent short circuiting. The modes of influent entry during the study were: (i) entry at the bottom of the reservoir, (ii) discharge 0.61 m above the reservoir water level, (iii) discharge 0.61 m above the reservoir water level using a spray attachment and (iv) discharge 0.61 m above the reservoir level through a venturi apparatus to add air to the stream. In addition to the four tests with varied types of influent entry, a coarse bubble aeration system was added to entry types (i) and (ii). Influent flowrates of 1.3 m^3/day, 3.3 m^3/day and 4.9 m^3/day (theoretical detention times of approximately 8, 12, and 30 hr, respectively) were tested for all operating conditions. Duplicate influent and effluent water samples were taken at 0.5 hr intervals over a 3 hr period and were taken only after the system had been operated for at least 3 detention times at each condition.

A dye test was conducted to determine the hydraulic detention time of the low technology tank. Rhodamine B was added to the tank, after it had filled with raw water, and the contents were mixed with a large paddle to achieve homogeneity. Then the flow of water through the system was started and a time = 0 sample of the effluent and tank contents was taken. The effluent dye concentrations were measured, using a Turner Model C10-005 fluorometer (Sunnyvale, CA), after 0.25, 0.5, 0.75, 0.88, 1 and 1.5 theoretical detention times. Studies were conducted using the 1.3 m^3/day and 4.9 m^3/day flow controllers.

The spray attachment (entry mode iii) was a standard garden hose screw-type adjustable nozzle. The diameter of the spray was set at

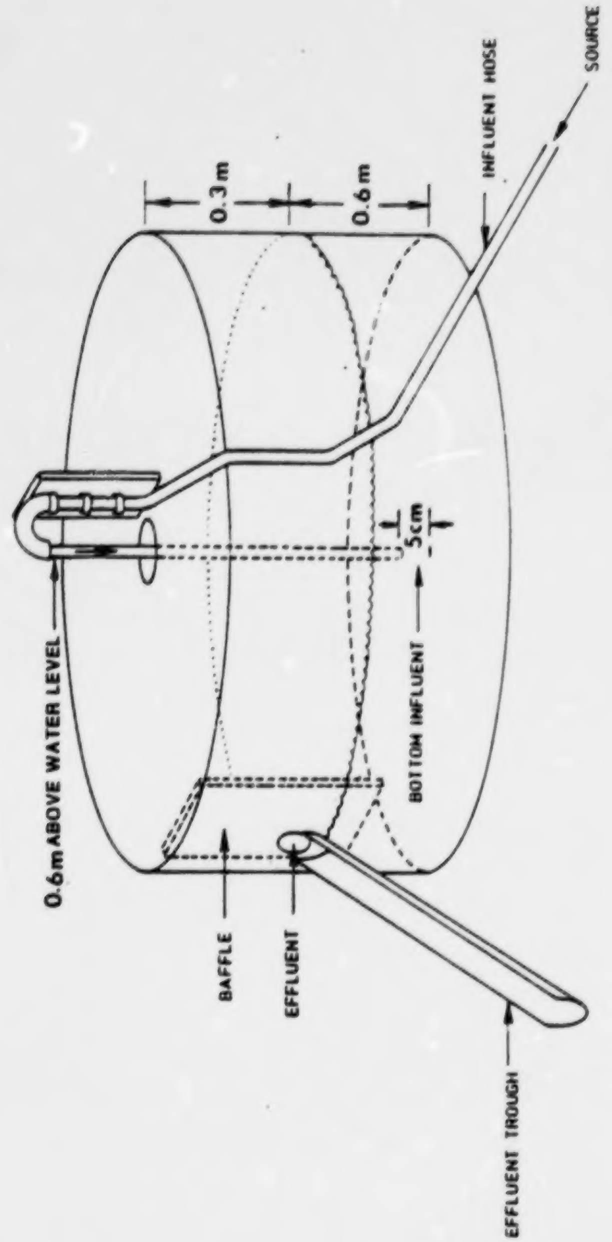

0.6m ABOVE WATER LEVEL
BAFFLE
EFFLUENT
EFFLUENT TROUGH
BOTTOM INFLUENT
5cm
0.3m
0.6m
INFLUENT HOSE
SOURCE

Figure 94. Schematic of the pilot-scale atmospheric storage tank. Derry, NH.

approximately 15 cm when it reached the water's surface. At the low flow of 1.3 m^3/day, a fine spray could not be achieved and thus data are not available for that condition. In tests where the coarse bubble aeration system was used, a laboratory-made apparatus was added to the reservoir. Air was supplied by a small laboratory air pump (capacity = 0.031 m^3/min, max. pressure = 414 kPa) and pumped through 0.64 cm (I.D.) clear plastic tubing. The main air line to the storage reservoir was subdivided into four 81 cm symmetrical radial arms using plastic connectors. The radial arms were plugged at the terminal ends with small corks. Small holes were pressed in the wall of the radial arm tubing, every 10 cm, using a steel-plated 11 mm long thumbtack which made 635 µm (I.D.) holes. The bubbles emanating from the tubing appeared to be approximately 4.8 mm diameter and were produced at an average rate of 2 to 3 per second from each hole. The venturi device (entry mode iv) used was designed for a laboratory faucet. The lowest flow accommodated by commercially available venturi devices was 27 m^3/day and therefore, could not be retrofit to the reservoir system.

RESULTS AND DISCUSSION

Loss in a Distribution Systems

The results obtained from the distribution system evaluations are summarized in Table 23. The overall loss during distribution was low (0 to 18.8%), over a distance of 497 m (hydraulic detention time 0.7 to 17.9 min). The greatest reduction (15.6%) in radon activity occurred at the home located furthest from the pumphouse and therefore where the water had the longest hydraulic detention time in the system. The differences observed, however, were not always statistically significant (Table 24).

During the evaluation with the untreated water, faucets were allowed to flush for several minutes, but it is possible that at site KO an insufficient time was allowed to completely clear the piping and the very small home storage tank. During some sampling, especially at sites KO and CU, it was very difficult to sample from the faucet without entraining air in the sampling syringe, even when using the universal hose connector suggested by EPA (1978).

The loss of radon due to decay alone would be minimal in the distribution system at Mont Vernon because of the short detention time (0.7 to 17.9 min) at the 82 m^3/day water flowrate. The community's daily flowrate is usually a minimum of 27 m^3/day, with a daily maximum of 82 m^3/day. At these flow conditions the loss of radon from decay would range from <0.01% to 0.67% in the homes sampled. Although the sampling techniques may have accounted for some of the observed radon removal, the mobile home park is known to have many leaks in its distribution system which may account for the radon loss.

The data suggest that flow in a small community distribution system (3.8-5.1 cm diameter pipes) at distances up to 497 m and detention times of 18 min or less will not significantly reduce the radon activity of the raw water. It is doubtful that this mechanism would be useful in lowering the radon levels to meet proposed standards unless a very small decrease was needed (10 to 20%). However, data were collected at Mont Vernon during periods of high

TABLE 23. LOSS IN THE DISTRIBUTION SYSTEM

Site	Distance (m)	Hydraulic Detention Time* (min)	Percent Radon Removal		
			RUN 1[+]	RUN 2	RUN 3
MA	35	0.7-1.2	0	0	2.1
SI	52	1.0-1.8	**	**	N/A
KO	100	2.0-3.6	10.8	7.3	++
CU	220	4.4-7.8	4.1	9.2	11.8
LA	497	10.0-17.9	15.6	**	7.5

[*] Hydraulic detention time range for 82 m^3/day water flowrate assuming 3.8 cm and 5.1 cm diameter piping.

[+] Runs 1 and 2 with GAC-treated water. Run 3 with untreated water.

[**] Radon level slightly higher than influent.

N/A Homeowners not available during sampling.

[++] Invalid sampling procedure (inadequate flushing or sample aeration).

TABLE 24. STATISTICAL COMPARISON* OF RADON REMOVALS WITH TIME IN A DISTRIBUTION SYSTEM

Site	Hydraulic Detention Time** (min)	Statistical Comparisons vs. System Influent+		
		RUN 1++	RUN 2++	RUN 3++
MA	0.7-1.2	INF = MA (α=0.05&0.10)	INF = MA (α=0.05&0.10)	INF = MA (α=0.05&0.10)
SI	1.0-1.8	INF = SI (α=0.05&0.10)	INF = SI (α=0.05) INF>SI (α=0.10)	N/A
KO	2.0-3.6	INF > KO (α=0.05&0.01)	INF > KO (α=0.05) INF = KO (α=0.01)	N/A
CU	4.4-7.8	INF > CU (α=0.05) INF = CU (α=0.01)	INF > CU (α=0.05) INF = CU (α=0.01)	INF > CU (α=0.05&0.01)
LA	10.0-17.9	INF > LA (α=0.05&0.01)	INF = LA (α=0.05) INF < LA (α=0.10)	INF > LA (α=0.05) INF = LA (α=0.01)

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* As assessed by Student's t test. α: significance level.

+ INF = Radon activity entering the water distribution system.

** Hydraulic detention time range for 82 m^3/day water flowrate, assuming 3.8 cm and 5.1 cm diameter piping.

++ Runs 1 and 2 = GAC-treated water, Run 3 = untreated water.

N/A Invalid sampling procedure or homeowner not available.

flow and thus probably represented the shortest detention time in the system. Though small reductions in radon are anticipated in short distribution systems, the removal rate would vary with water usage. Removal of radon could be higher if the water was retained in the system for a period of time, however, that condition could not be relied upon to consistently deliver water of acceptable quality.

Loss During Storage

A series of dye studies were completed on the flow-through reservoir. Tests were conducted to determine if the tank was well mixed. The data indicate that the hydraulic detention time at the high flowrate (Q = 4.5-4.8 m^3/day) was very close to the theoretical time of 8 hr (8.27 ± 0.40 hr) Figure 95). There was no stratification within the tank as a function of T°C or dye concentration at this flowrate. At the low flow condition (Q = 1.2-1.4 m^3/day), however, the calculated detention time from the dye study was 23.3 ± 6.72 hr, as contrasted to the theoretical time of 30 hr (Figure 96). The detention time for the three separate dye studies ranged from 18.9 to 30.3 hr. Though the water volume of the tank remained constant (2,043 L), the T°C and dye concentrations varied with depth in the tank, indicating that flow was short circuiting across the surface of the tank. In addition, the 1.3 m^3/day flow controller tended to clog with fine particles even when a sediment trap was used. As a result, the flow slowly decreased with time. Possibly account for the larger detention time during the second trial. This implies that the data obtained during subsequent studies for the reservoir using the 1.3 m^3/day flow controller probably underestimated the removal attainable at a true detention time of 30 hr. A dye study was not run using the 3.3 m^3/day flow controller, however, it was observed during the loss studies to respond similarly to the 4.9 m^3/day controller therefore, it is assumed that no problems with clogging or short-circuiting occurred at this flow condition.

The data collected during the pilot-scale flow-through reservoir studies at Derry are summarized in Figures 97, 98 and 99. The radon removals achieved at the different detention times were significantly different for all conditions ($\alpha = 0.05$ & 0.01, StT) (i.e., removal always significantly higher with increased detention time) except for the venturi. However, results from the different flowrates should not be directly compared because of the potential problems with short-circuiting at the 1.3 m^3/day flowrate. The reduced removals with the venturi may have been a function of the fact it was designed for laboratory use as an aspirator. In addition, the flowrates tested were probably too low to generate good venturi action (aeration), preventing better removals from being observed in the venturi runs. Testing should be performed at higher flowrates with a commercially-available device to thoroughly evaluate the venturi's ability to enhance radon removal.

Losses for the bottom entry of the influent into the reservoir were very similar to the removals expected as a result of decay and volatilization of radon. However, in all cases where the influent had a 0.61 m free fall upon entry into the tank, radon removals were much greater than those predicted from decay alone or bottom entry without aeration. (N.B., statistical analyses of the data were not warranted because the influent radon activity changed among some of the experiments at a given flowrate from 58,641 to

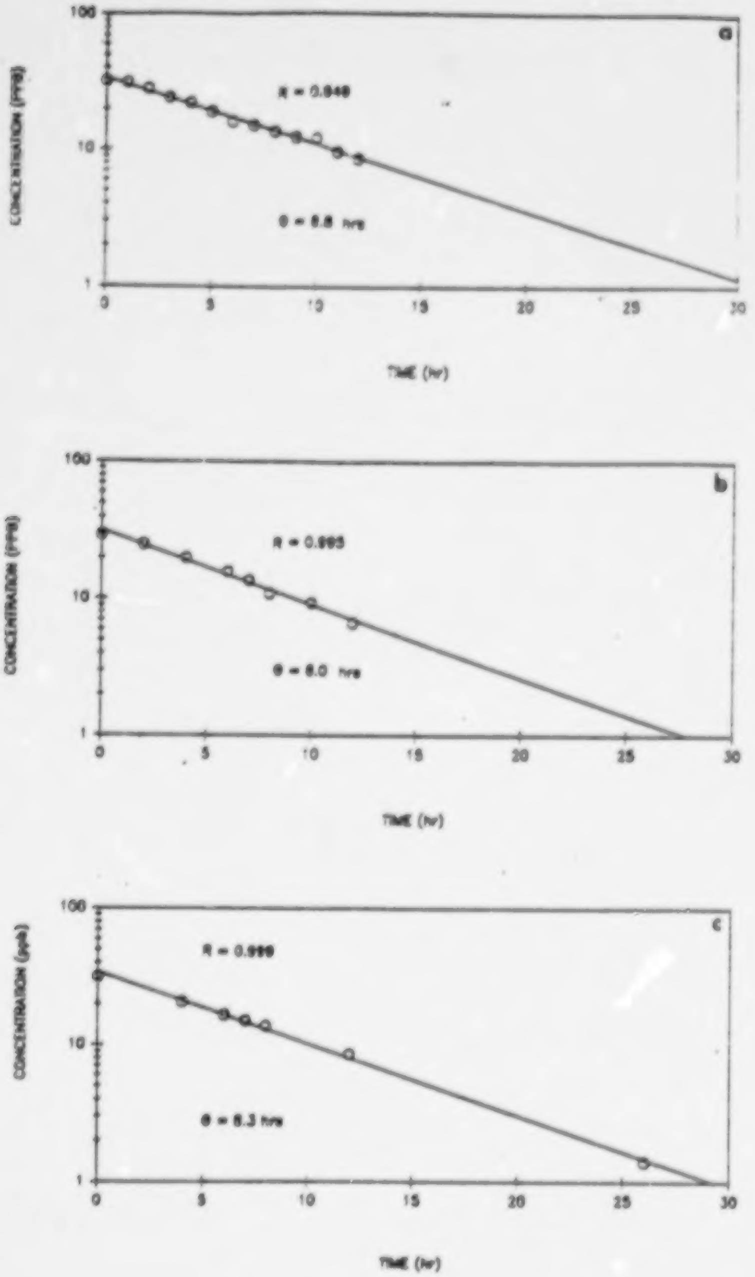

Figure 95. Low technology alternatives - Derry, NH. Dye concentration in pilot-scale atmospheric storage tank with free fall as a function of time. Runs 1 (a), 2 (b), and 3 (c). (Water flowrate = 4.9 m^3/day).

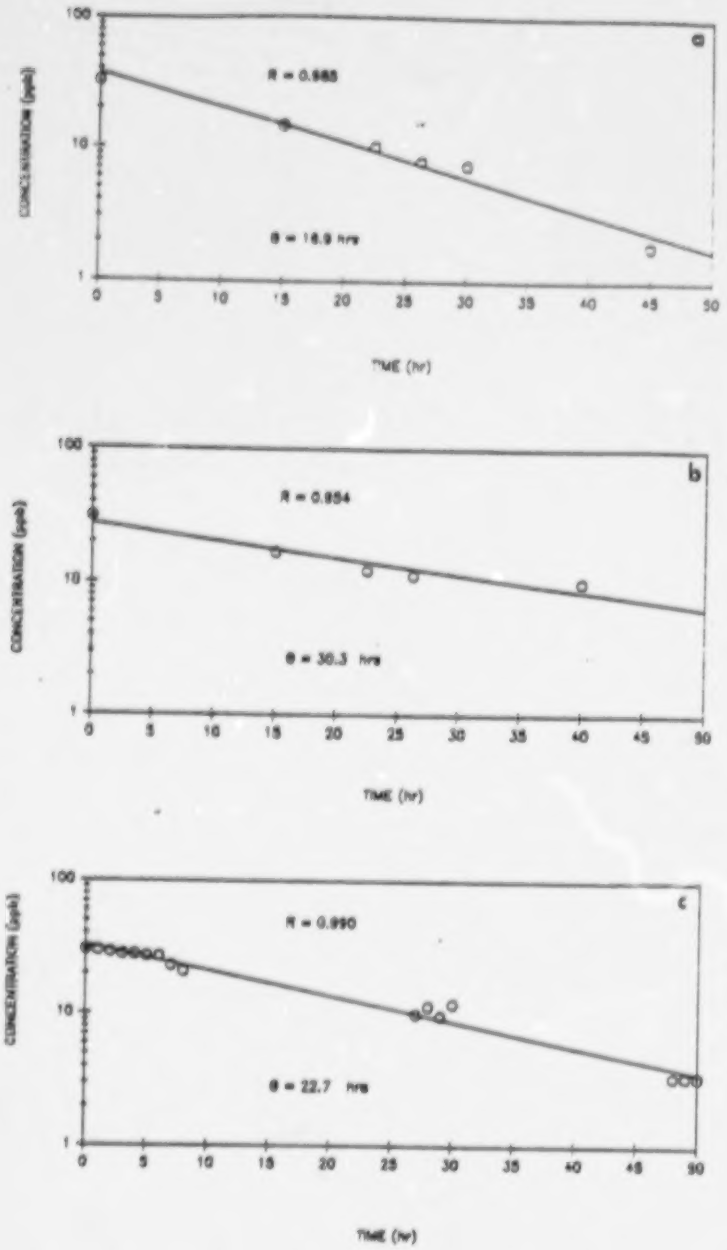

Figure 96. Low technology alternatives - Derry, NH. Dye concentrations in the pilot-scale atmospheric storage tank with free fall as a function of time. Runs 1 (a), 2 (b) and 3 (c). (Water flowrate = 4.9 m^3/day).

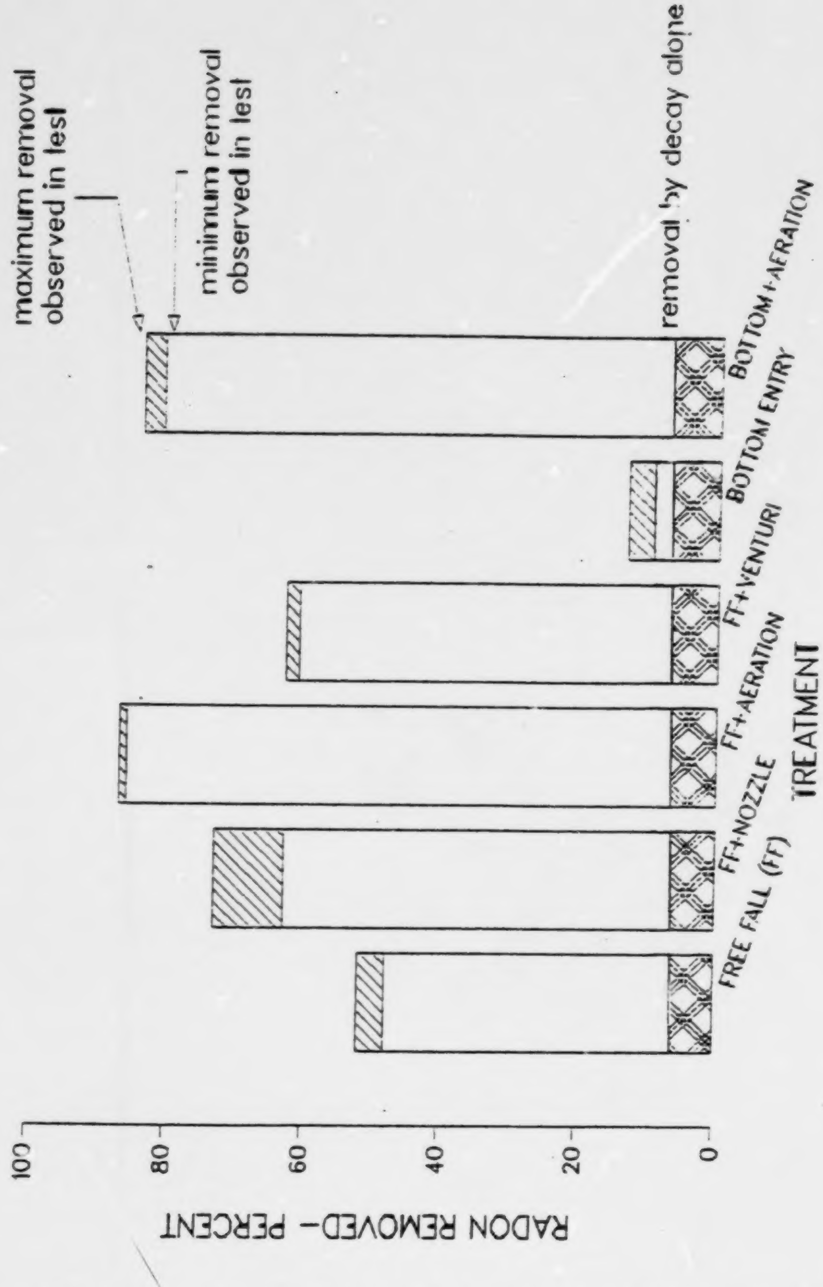

Figure 97. Low technology alternatives - Derry, NH. Percent radon removed for different conditions. (Water flowrate = 4.9 m^3/day).

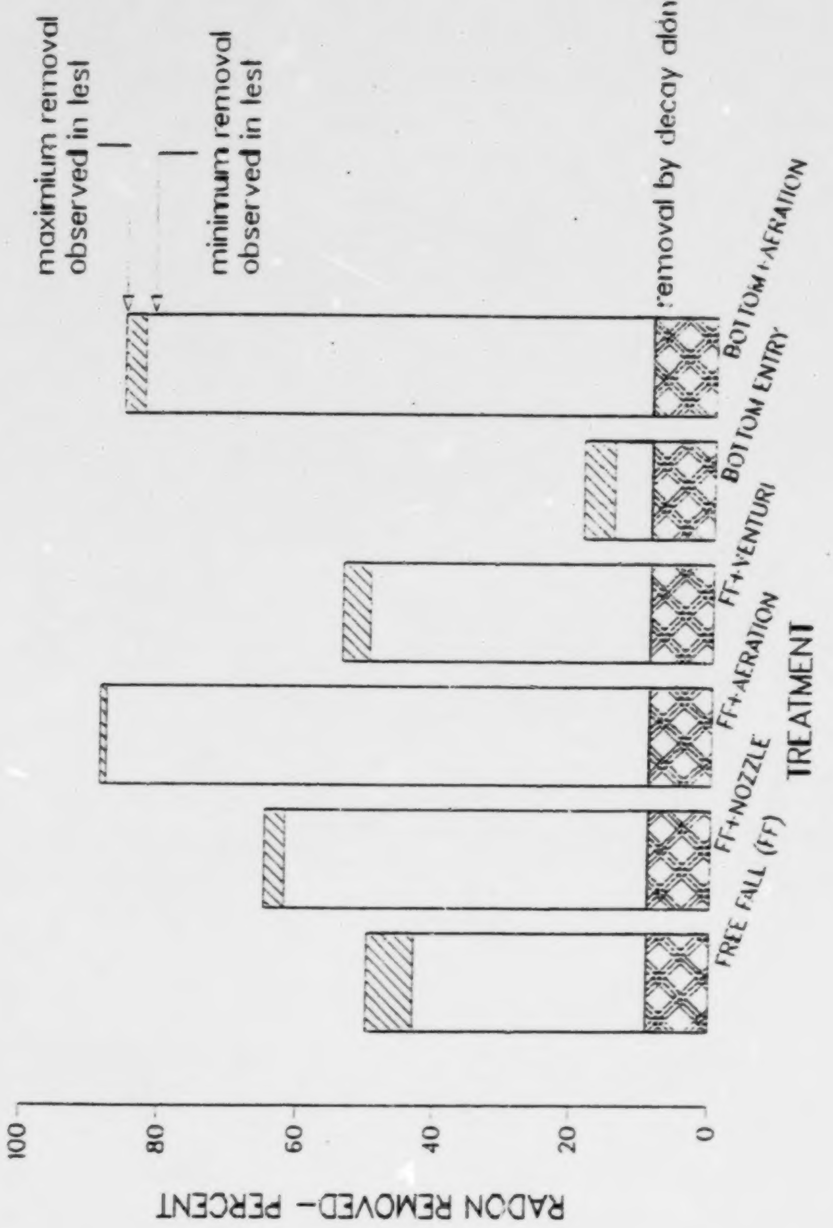

Figure 98. Low technology alternatives - Derry, NH. Percent radon removed for different conditions (Water flowrate = 3.3 m³/day).

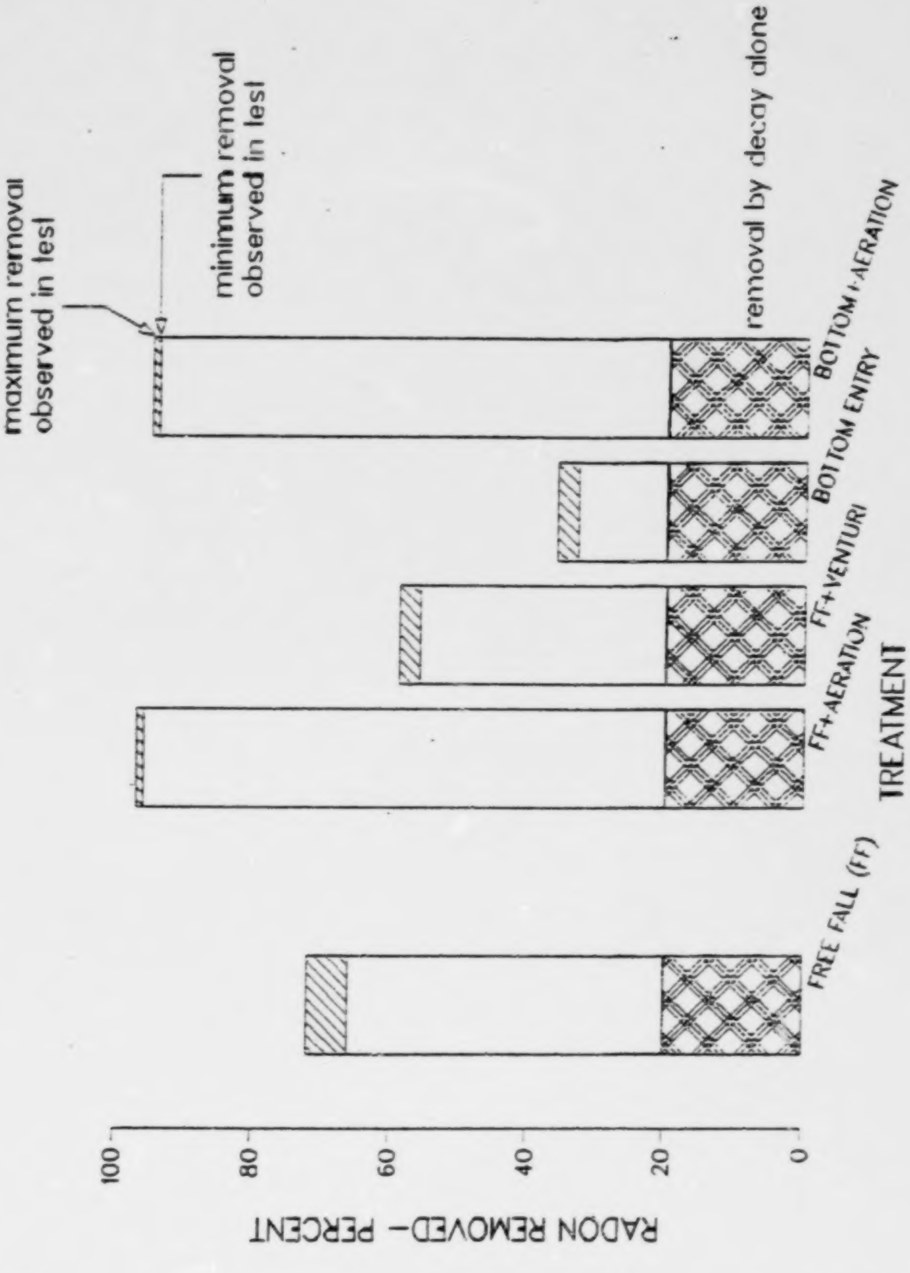

Figure 99. Low technology alternatives - Derry, NH. Percent radon removed for different conditions (Water flowrate = 1.3 m^3/day).

95,704 pCi/L.) While removals with the 0.61 m free fall entry alone were 50% (3.3 and 4.9 m^3/day) to 70% (1.3 m^3/day), they were enhanced by the use of the spray nozzle (60-70%, 3.3 to 4.9 m^3/day) and the coarse bubble aeration system (86-89%, 3.3 to 4.9 m^3/day; 96%, 1.3 m^3/day). The radon removal was also high using the coarse bubble aeration system in conjunction with the bottom entry to the reservoir; removals ranged from 83% (4.9 m^3/day) to 95% (1.3 m^3/day). The data collected in this phase of the study showed that addition of a coarse bubble aeration system to a storage tank may be very effective for radon reduction and might be easily applied in small communities which have existing atmospheric storage tanks. However, pilot scale testing would be needed because sidewall depth could affect removals. In the reservoir tested, the sidewall depth was only 0.61 m. Existing storage tanks may be much deeper causing poorer mass transfer (i.e., bubbles to be saturated with radon before they reach the surface) thus requiring greater air flowrates (A:W ratios) to achieve effective radon removal.

Shortly after our pilot-scale study at Derry was completed, the local water authority decided to add a coarse bubble aeration system to the existing atmospheric storage tank. Two 6 m long PVC pipes (5 cm I.D.) were installed 81 cm apart and 46 cm off the bottom of the tanks. The bottom part of each pipe contained a series of 3 mm diameter holes drilled every 5 cm for the first 4.6 m of length and at 2.5 cm intervals for the remaining 1.5 m. Raw water was pumped from the wells to the terminal end of the tank (6.7 m) before exiting the influent pipe a minimum of 0.30 m above the water's surface. A compressor delivered 0.57 m^3/min of air to the tank through the perforated pipes and was run continuously. The average detention time of the water in the tank was 5.3 hr. Two 15 cm diameter vent pipes were welded on top of the tank for ventilation. Radon removal efficiencies ranged from 80 to 88%, the latter occurring after approximately 6-8 hr of contact (overnight).

Several potential problems may arise with these kinds of coarse bubble aeration techniques. No gas phase monitoring was conducted during the study so the fate of the radon release into the ambient air remains unknown. However, based on the diffused bubble and packed tower aeration data, this may be a concern and should be monitored. In addition, water supplies which contain significant amounts of reduced iron and manganese may experience problems with precipitate formation and deposition in the storage tank and/or the distribution system.

It is important to note that in the case of this water supply the reductions of 80% to 96% observed for the coarse bubble aeration system would not be sufficient treatment to attain the effluent activity of 200-1,000 pCi/L which may be required by the MCL (influent activity = 58,641 to 95,704 pCi/L). In addition, extrapolation of the performance data to design systems which can achieve these required levels for this water supply is not advisable based on the importance of mass transfer limitations found in the diffused bubble and packed tower aeration studies at low radon activities. Additional pilot-scale testing at low radon activities would be required to determine if the coarse bubble aeration system could attain effluent activities of 200-1,000 pCi/L at Derry, NH.

ECONOMICS

Several low technology systems were evaluated for radon removal in small communities. However, since the major costs of these low technology systems involve retrofitting and system specific designs, it is not possible to provide a detailed estimate of these system costs. Therefore, a simple cost example of one type of low technology treatment will be presented for illustrative purposes.

The cost example is based on the low technology coarse bubble aeration system used by the water authority at Derry, NH. Cost estimates for this low technology system are shown in Table 25. The techniques used to estimate the costs of the low technology system are analogous to those described in Section 4 with the following exceptions: 1) support equipment costs were minor and thus combined with process equipment costs; 2) indirect costs were considered negligible and thus omitted from the analysis; 3) O&M costs were based solely on power and maintenance costs; and 4) pretreatment costs were not considered since it was assumed that construction of a sophisticated pretreatment system for iron and manganese would defeat the purpose of using a low technology system for radon removal. Based on these assumptions, the total production cost estimate of retrofitting the full-scale low technology system used at Derry, NH was \$0.15/1,000 gallons. It should be noted that the cost estimate assumes no treatment of the radon contaminated vent gas other than discharging at the average home roof line (i.e., 10.7 m) is required.

TABLE 25. COST ESTIMATE FOR LOW TECHNOLOGY TREATMENT AT DERRY, NH

Item	Total Cost (2nd Quarter 1989 dollars)*
CAPITAL COSTS	
12.2 m (5.08 cm I.D.) PVC aeration system (Assembled and Installed)	$ 208
0.5 Hp air compressor	630
15.2 cm diameter air vent system (Assembled and Installed)	446
TOTAL CAPITAL COST	$ 1,284
AMORTIZED CAPITAL COST (Annual)	$ 141
ANNUAL O&M COSTS	
Power	332+
Maintenance	
Mechanical	63
Non-mechanical	26
TOTAL ANNUAL O&M COST	$ 421
TOTAL ANNUAL COST	$ 562
TOTAL PRODUCTION COST (Design flow 37.85 m^3/day)	$0.15/1,000 gallons

*Based on ENR Construction Cost Index, Base Year 1967 = 100.

+Includes required input power of 0.5 Hp for the compressor, operating period of 24 hr/day, and an electric rate of $0.10157/kW-hr.

SECTION 8

EVALUATION OF THE LIQUID SCINTILLATION TECHNIQUE FOR RADON ANALYSIS

INTRODUCTION

As part of the maximum contaminant level (MCL) regulation for radon in drinking water, EPA must recommend sampling and analytical techniques to insure uniformity and comparability in monitoring and compliance. At a minimum, the method(s) recommended must be reliable, available and have an acceptable practical quantification level. In addition, easy operation and moderate cost are desirable characteristics. There have been several techniques developed to measure radon in water. In the Lucas cell method (Lucas, 1957), radon is de-emanated from a water sample into a zinc-sulfide phosphor-coated chamber where the alpha scintillations occurring as a result of radon and its progeny's radioactive decay are counted by a photomultiplier detection system. Though the procedure is labor and equipment intensive, it is the most sensitive of all of the radon techniques available (0.2 pCi/L for a 60 mL sample) (Yang, 1987). Lucas (1964) developed a faster method, later modified by Irfam and Read (1981), which uses gamma spectroscopy for monitoring radiation from the radon progeny, polonium-214 and bismuth-214. The procedure involves minimal sample preparation, but requires large sample volumes (1 L) to have a detection limit in the range of 10-100 pCi/L. Alpha track detector cups, without tops, have also been used for water storage tanks. Radon released from the water is trapped in the inverted cup and energy from alpha decay causes sub-microscopic tracks in the plastic material in the cup. The tracks are made visible by etching the plastic in a caustic soda solution. The number of tracks is counted microscopically. However, the method is difficult to calibrate because it depends on how often the toilet is flushed (P.B. Hahn, USEPA, Environmental Monitoring System Laboratory - Las Vegas, personal communication, 1989). Orme and Eaton (1986), adapted a method used in oceanography, where radon, purged from water, is concentrated on activated carbon and subsequently desorbed into a Lucas cell. The method has a minimum detection limit of 2.0 pCi/L.

Perhaps the most promising method for widespread monitoring because of its reliability, simplicity and speed (~1 min preparation time) is the liquid scintillation technique originally developed by Pritchard and Gesell (1977). In this procedure, a water sample (5 or 10 mL) is injected into a glass vial containing a liquid scintillation cocktail. The cocktail consists of a high molecular weight organic compound (fluor) and a solvent. The solvent originally used was toluene because radon is 12.7 times more soluble in toluene than in water and therefore can be readily extracted from a water sample into the cocktail (Pritchard and Gesell, 1977). A period of 4 hr after injection is required before the samples can be analyzed to allow secular equilibrium to develop between radon and its short-lived progeny. When alpha and beta particles created during the decay of these radionuclides strike the fluor molecules in the cocktail, they emit light pulses which are counted by photomultipliers located in the automated liquid scintillation counter. The decay series of radon-222 through polonium-214 yields three alpha particles (5.49, 6.0, and 7.69 MeV) and two beta particles with end points ranging from 0.69 to 3.26 MeV. Liquid scintillation techniques based on measurement of these kinds of energetic particles are approximately 100% efficient. Detection limits as low as 10 pCi/L (40 min counting time) have been reported for this technique (Pritchard and Gesell, 1977). However, recent research by Hahn (USEPA Environmental Monitoring Systems Laboratory - Las Vegas, personal communication, 1989) indicates that in order to have counting uncertainties (2σ) of $\leq$ 10% when measuring samples with low activity ($\leq$ 500 pCi/L) counting times >> 100 min are required (background counting rate ~100 cpm). If the MCL for radon in drinking water is set in the range of 500 to 1,000 pCi/L, counting times of approximately 60 to 20 min, respectively, would be required to achieve a 10% counting uncertainty (2σ) (P.B. Hahn, USEPA Environmental Monitoring Systems Laboratory - Las Vegas, personal communication, 1989).

The liquid scintillation technique is the only method allowing relatively rapid processing of samples and requiring small sample volumes. These advantages are important because compliance with the radon regulation will probably necessitate analyzing millions of water samples each year. The method has one disadvantage in that it lacks specificity. It will detect any alpha or high energy beta-emitting radionuclide soluble in the liquid scintillation cocktail. However, this drawback is considered insignificant in most groundwater supplies because radon is the primary radioactive contaminant soluble in the two-phase cocktails used.

EPA (1978) has used the liquid scintillation technique in its national monitoring programs to determine the distribution of radon concentrations in water supplies. Their procedures specify mineral oil as the cocktail solvent because toluene cannot be mailed without special handling and can present a problem with respect to laboratory safety and disposal. In addition, EPA collects samples in volatile organics bottles (VOA) and processes them in the laboratory (i.e., injection into the vial containing the cocktail). Though the EPA procedure adds an extra step in processing, it avoids widespread distribution of syringes.

There are several ways to calculate the radon present in the original sample. EPA (1978) uses the formula:

$$A = \frac{(C_S - C_B)\ (1000\ \text{mL/L})}{(E)\ (V)\ (D)} \qquad \text{(eq. 13)}$$

where:

A = radon activity in original water sample (pCi/L)

C_S = sample counts per minute (cpm)

C_B = background obtained from counting an equivalent volume of laboratory grade water in the same scintillation cocktail (cpm)

E = efficiency factor (cpm/pCi)

V = volume of sample analyzed (mL)

D = decay correction = $e^{-0.693T/t_{1/2}}$

T = time from sample collection to midpoint of counting time (days)

$t_{1/2}$ = half-life of radon-222 (3.82 days)

The efficiency factor (E) is based on the cpm obtained from counting a standard containing a known amount of radon (dpm or pCi). The standard is prepared by dissolving a known amount of radium-226, the long-lived parent of radon-222, in water. After a period of 20-25 days (i.e., 6 half-lives), the radium-226 is in secular equilibrium with radon so that a constant amount of radon-222 is always present in the vial.

A series of experiments was conducted to evaluate how certain factors affected the liquid scintillation technique results (i.e., radon concentration and variability). Factors investigated included: (i) sampling in the field with a syringe and immediate injection into the liquid scintillation vial versus collection in a VOA bottle and subsequent laboratory processing (Sampling Experiment); (ii) storage in VOA bottles (Storage Experiment); (iii) groundwater source, sample preparation and instrumentation variability (Variability Experiment); (iv) different liquid scintillation cocktails (Cocktail Experiment); and (v) extraction of radon into the cocktail (Extraction Experiment).

METHODS AND MATERIALS

Samples from all of the experiments were collected and analyzed according to the procedure outlined in Section 3 with specific modifications noted below for each experiment. They were all counted for 10 min using a Beckman LS 7000 Liquid Scintillation Counter (Fullerton, CA) with windows set at 0-397 (channel 1) and 397-940 (channel 2). Counts from both channels were used to calculate radon activities. A correction for interference was made for each sample using an internal cesium-137 source. This procedure is called the quench determination and is expressed as an H number (LS 7000 Program Number 6 - Appendix B). A Beckman tritium (hydrogen-3) instrument calibration standard, a radium-226 standard and a field blank (made from double deionized water (DDI)) were counted at the beginning of each analytical run. Duplicate analyses were performed every tenth sample. All glassware used in the experiments was cleaned in warm, soapy tapwater, rinsed three times with deionized water (DI), soaked for 1 hr in 10% Extran (VWR Scientific; Boston, MA), rinsed three times with DI, rinsed once with DDI and air dried.

The radium stock solution (117.5 pCi/mL) was prepared from a radium-226 (4.45 nCi/g) standard supplied in a glass ampule by the USEPA Environmental Monitoring Systems Laboratory (Las Vegas, NV).

The contents of the ampule were emptied into a tared beaker and weighed. They were subsequently transferred to a volumetric flask with several rinses of 0.05M HNO_3 and diluted to 200 mL with 0.05M HNO_3. This 200 mL dilution was used as the stock radium-226 solution. Each radium-226 standard was prepared by pipetting 5 mL of the stock and 5 mL of 0.05M HNO_3 into a 20 mL low absorbance glass liquid scintillation vial to achieve a final activity of 588 pCi. The standards were purged of radon by bubbling nitrogen gas through them for 20 min. 10 mL of liquid scintillation cocktail was slowly added to each vial before capping. Additional details of the radon analytical technique are given in Section 3.

Sampling Experiment

Radon samples were collected from two homes: one in Deerfield, NH (radon activity = 46,600 - 57,500 pCi/L) and one in Lee, NH (radon activity = 1,800-2,700 pCi/L). At each site, aerators on the faucets were removed before sampling and 18 samples were collected: 6 were obtained in amber VOA bottles fitted with teflon septa using the universal faucet connector (EPA, 1978), 6 were collected in amber VOA bottles using free falling water (0.15-0.30 m) directly from the faucet, and 6 were collected from the faucet connector using a 10 mL gas-tight glass syringe fitted with a 13 gauge needle and were injected directly into vials containing a cocktail of Liquifluor (40.3 mL/L) (New England Nuclear; Boston, MA) in a balance of liquid scintillation grade toluene (toluene-based cocktail). For all of the sampling techniques, the water flowrate was set at 927 mL/min at Lee and 800 mL/min at Deerfield. Samples were collected after the water had run for 3 min. The order of sampling was integrated, so that immediately after a free fall sample was taken, one was collected using the faucet connector, followed by one using the syringe. This alternation of sampling methods (free fall, faucet connector, syringe) throughout the collection period avoided confounding (time effects) in the results. Four field blanks of DDI (2 VOA bottles, 2 direct syringe) were collected at the end of each sampling. The entire collection period at a given site was performed within 15 min. After filling, VOA bottles were checked to ensure that air bubbles were not present. Once in the laboratory, 10 mL water samples were removed from each VOA bottle using a 10 mL gas-tight glass syringe and were injected into a toluene-based cocktail. All samples were then analyzed in the liquid scintillation counter as outlined in Section 3.

Storage Experiment

Samples were collected from an abandoned municipal ground water supply in Derry, NH (radon activity = 77,477 ± 6,512 pCi/L) using 60 mL amber VOA bottles filled using the universal faucet connector. A total of 24 water samples and 11 DDI field blanks were collected. In addition, 7 samples were taken using the direct syringe technique. Immediately, after all of the samples were collected (10 min), aliquots from 2 sample VOA bottles and 1 blank VOA bottle were taken and added to scintillation vials containing the toluene-based cocktail. After returning to the laboratory, these 3 samples, as well as the 7 direct syringe technique samples were counted immediately. All other sample and blank VOA bottles were returned to the laboratory and stored at room temperature (18-22°C). Two sample VOA bottles and one blank VOA bottle were processed and analyzed after storage periods of 0.4, 1.0, 3.0, 5.0, 7.0, 9.0, 11.0, 13.0, 15.0, 17.0 and 21.0 days.

Variability Experiment

A hierarchical experiment (Box et al., 1978) was conducted to determine the variation in radon activity associated with a groundwater source, sample preparation and instrumentation. Fourteen samples were collected at the Derry, NH site: 7 using the 60 mL amber VOA bottles and the universal faucet connector with subsequent laboratory processing and 7 using the universal faucet connector and the direct syringe technique. Two subsamples were processed from each VOA bottle. A toluene-based cocktail was used for all samples. The contents of the vials were analyzed in the liquid scintillation counter on 2 consecutive days according to the procedure outlined in Section 3.

Cocktail Experiment

Six radium-226 standards were prepared using a toluene-based cocktail; 3 using a cocktail containing Liquifluor (40.3 mL/L) in a balance of mineral oil (New England Nuclear; Boston, MA) and 3 using an Opti-Fluor O cocktail (Packard; Sterling, VA). All standards were prepared on the same day from the same radium-226 stock solution and were analyzed each day for a period of 29 days, then every 2-3 days for 2 weeks and weekly thereafter for a period of 7 months. In addition, 6 water samples were collected using the direct syringe technique with the universal faucet connector over a period of 20 min using each cocktail at both the Derry and Lee, NH sites. As in the Sampling Experiment, the order of sampling was integrated. Immediately after a toluene-based cocktail sample was taken, one was collected in the mineral oil-based cocktail, followed by one in the Opti-Fluor O. This alternation of cocktails used throughout the sample collection period avoided confounding in the results. All standards and samples were analyzed as outlined in Section 3.

Extraction Experiment

Three identical sets of four radium-226 standards were prepared using each cocktail (toluene-based, mineral oil-based and Opti-Fluor O): Set A was shaken before every analysis; Set B was shaken only on the day the standard was prepared; and Set C was never shaken. In addition, 12 samples were collected using the direct syringe technique with the universal faucet connector over a period of 15 min at both the Derry and Lee, NH sites. Six samples of each cocktail from each site were shaken for 1 min immediately after collection. All samples and standards were analyzed according to the method outlined in Section 3.

RESULTS AND DISCUSSION

Sampling Experiment

Radon samples are usually collected in the field in VOA vials and sent to the laboratory for subsequent processing. This avoids distribution of syringes and volatile scintillation cocktails, and problems with collection of bubbles within the syringe. EPA (1986) recommends filling the bottles with a universal connector attached to a faucet without an aerator. In the sampling experiment, the direct collection of samples using a syringe from a faucet fitted with the universal connector was compared with collection in VOA bottles filled directly from the faucet (free fall) or filled using the universal connector.

The results of samples collected at the Lee and Deerfield, NH sites are presented in Table 26. At the Lee site, analysis of variance (ANOVA) showed that the direct syringe technique yielded significantly higher activity than the free fall and universal connector techniques at $\alpha = 0.05$, but not at $\alpha = 0.02$. There was no significant difference between either VOA sampling method ($\alpha = 0.05$ and 0.10). At the Deerfield site, none of the three methods of sample collection were significantly different at $\alpha = 0.05$ or 0.1.

Numerically, the direct syringe technique always yielded the highest activities of the three methods, while the VOA bottle filled with free falling water yielded the lowest. This is not surprising as less sampling handling prior to injection into the scintillation vial and less agitation during sampling should result in less loss of radon. However, though the VOA collection techniques involve extra handling of the sample, they can produce results statistically similar to the syringe method.

The direct syringe technique may be preferred by researchers because it has the potential to yield results with the least radon loss and sample handling. Its use in compliance monitoring may be limited because of the costs and safety problems associated with widespread distribution of gas-tight glass syringes and volatile scintillation cocktails. In addition, samples taken with syringes must be drawn slowly to avoid cavitation and inclusion of air bubbles. Though plastic disposal syringes may be used, they are not designed for air-tight sampling. Thirteen gauge needles are large enough to limit these problems with air bubbles, but even experienced persons can still have problems completely eliminating them.

Sample collection with the free falling water must be done carefully at low water flowrates (800 to 927 mL/min was used) to insure results comparable to the direct syringe method. Splashing water into the bottles at substantially higher water flows would probably yield markedly lower results. The proper sampling techniques would need to be clearly outlined in any sampling procedure supplied with the VOA bottles. Distribution of universal faucet connectors would be cumbersome and increase the costs of sampling. However, their use insures collection of samples at lower water flowrates. As a result, the samples collected using the connector may be more consistently representative of the radon activity in a water supply and therefore more desirable for compliance monitoring.

TABLE 26. SAMPLING EXPERIMENT RADON ACTIVITIES

Site	Direct Syringe (pCi/L)	Universal Faucet Connector (pCi/L)	Free Fall (pCi/L)
Lee, NH	2,360	2,290	2,072
	2,492	2,578	2,338
	2,360	2,261	2,228
	2,440	2,105	2,415
	2,524	2,360	2,198
	2,468	2,314	2,207
$\bar{X} \pm S$*	2,486 ± 90	2,318 ± 154	2,243 ± 120
Deerfield, NH	53,349	54,378	46,655
	53,358	52,941	50,313
	54,107	51,252	54,533
	51,441	48,826	46,832
	52,220	51,505	47,985
	57,446	59,591	50,825
$\bar{X} \pm S$*	53,664 ± 2,084	53,082 ± 3,688	49,524 ± 3,008

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*S represents 1 standard deviation.

Storage Experiment

Radon has a relatively short half-life necessitating a minimal storage time, especially for samples with activities near the MCL. Most radon sampling, conducted to ensure compliance with the MCL, will involve collection in VOA bottles, transportation (often by mail) and laboratory processing. The storage experiment was designed to examine the effect of the length of time between collection and laboratory processing on the results obtained. Losses of radon in the VOA bottles are caused by radioactive decay and possible leakage. The effect of decay is easily predicted based on the half-life of radon and the time between sample collection and analysis. The extent of leakage is unknown.

The 7 samples collected initially using the direct syringe technique had a mean radon activity and standard deviation of 65,068 ± 2,941 pCi/L. These samples were used as a basis of comparison for all of the VOA samples. The radon activity measured in the samples held in the VOA bottles decreased exponentially over time as predicted by radioactive decay (Fig. 100). When the results were corrected for the decay which occurred between the time of collection and analysis, only 4 samples (3.0, 5.0, 13.0, 17.0 days) had activities which were significantly different ($\alpha = 0.05$, StT) from those of the direct syringe method. (At $\alpha = 0.02$, only 3.0, 5.0 and 13.0 days were significantly different.) For the 3.0, 5.0, 13.0 and 17.0 day samples, the loss of radon ranged from 8.5 to 16.5%. There was no correlation between the length of storage time and leakage, indicating that leakage was probably a random function of specific bottles and their assembly or variability in sampling rather than storage time (Fig. 101).

The data indicated that loss of radon from VOA bottles could be a factor in some cases, but the loss resulting from radioactive decay has the greatest potential impact on storage time. Within 4 days, 50% of the radon originally present will be lost due to decay. For samples containing high levels of radon, permissible storage times could be substantial provided that the amount remaining at the time of analysis is above the practical quantification level. (For example, a sample containing approximately 7,670 pCi/L could be held up to 10 days, even with a 20% loss due to leakage, and still contain 1,000 pCi/L). However, the amount of radon in a sample is often not known, so the maximum storage time adequate to obtain a valid measurement must be based on the MCL.

Variability Experiment

The total variability and relative contribution of various sources of error within a sampling and analytical procedure can be determined using a hierarchical experiment (Box et al., 1978). In the variability experiment, samples were collected from a single water source over a 20 min period in order to measure the percent relative standard deviation (% RSD) associated with variation in the groundwater source, sample preparation and instrumentation. The hierarchical experiment was conducted using (i) direct syringe sampling and (ii) collection in VOA bottles with subsequent laboratory processing. The variation due to sample preparation could not be separated

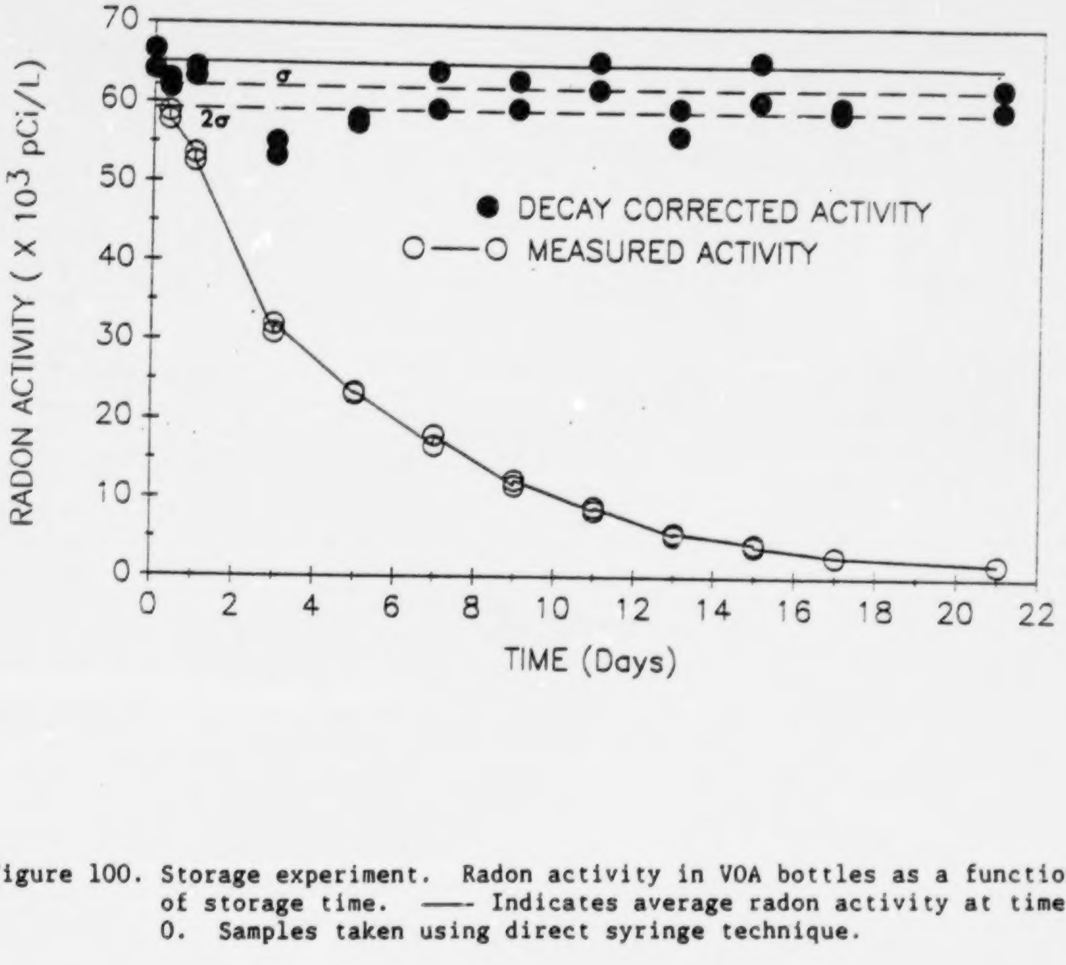

Figure 100. Storage experiment. Radon activity in VOA bottles as a function of storage time. —- Indicates average radon activity at time = 0. Samples taken using direct syringe technique.

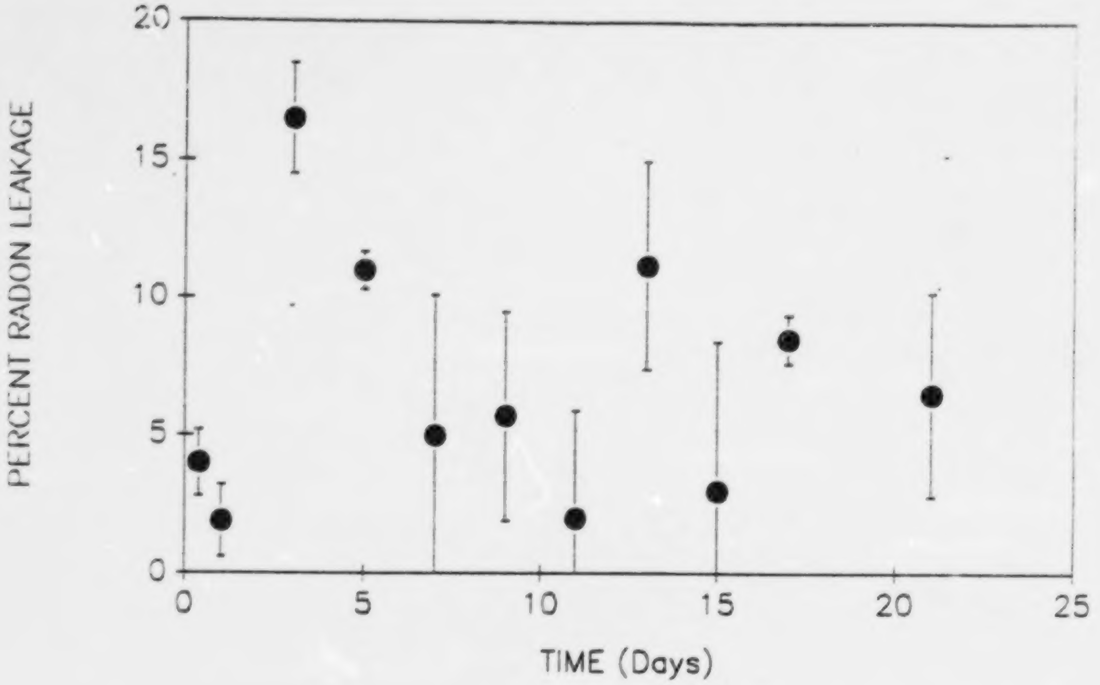

Figure 101. Storage experiment. Percent leakage of radon from VOA bottles as a function of storage time.

from the variation associated with the instrumentation for the direct syringe technique.

Table 27 shows the total variation and that associated with each phase of the process for both sampling methods. There was a significant difference ($\alpha = 0.01$ and 0.05, StT) between the mean radon content obtained by the two methods. The direct syringe technique yielded a significantly higher radon activity. This differed from the findings of the sampling experiment where the two methods yielded similar results. During the variability experiment, the water flowrates were higher which probably resulted in greater loss of radon when the VOA bottles were filled. However, as noted in the sampling section, the direct syringe procedure would require the widespread distribution of syringes which is not desirable and may offset its 5 to 6% higher activities.

The total variability associated with the two methods was not significantly different ($\alpha = 0.05$ and 0.10, F test). Total variation of 4 to 6% as a result of sample handling and instrument variation is not high considering the volatile nature of radon. For example, purgeable organics analysis for water may have standard deviations as high as 10 to 20% (EPA, 1984). In an interlaboratory collaborative study using sealed radium-226 standards, Whittaker et al. (1987) found within-laboratory average repeatability for the liquid scintillation techniques of 3.6 ± 3.0% at 95% confidence (18 laboratories). Their data includes variations due to sample preparation and instrumentation and agrees with the mean %RSD results shown in Table 27.

For many contaminants, the variability in the concentration in the environment represents the largest component of the standard deviation. For radon, very little variation was due to changes in the water source (0-1.6%) over the 20 min sampling period. This may not be true, however, for new wells or those sampled over much longer intervals (F.R. Hall, University of New Hampshire, Department of Earth Sciences, personal communication, 1988). Most of the variability in the direct syringe method (92.1% of total variance) was due to a combination of sample handling and instrumentation. In the VOA technique, the variation (as percent of total variance) due to sample preparation and instrumentation was 55.5% and 44.5%, respectively. Use of an air inlet needle when removing the sample from the VOA bottle and slow injection of the water from the syringe beneath the cocktail's surface are essential to having a small variation associated with sample handling. The air inlet needle prevents a vacuum from forming when the 10 ml sample is withdrawn from the VOA bottle. Tightly capping the scintillation vials and wiping their outside walls clean prior to placement in the counter are also important steps.

Cocktail Experiment

In the original liquid scintillation method developed by Pritchard and Gesell (1977), toluene was chosen as the cocktail solvent. The major problems associated with use of toluene-based cocktails are mailing restrictions, laboratory safety, its volatility and waste disposal. To avoid these problems, EPA (1978) prescribed mineral oil as the solvent for the cocktail

TABLE 27. RESULTS OF THE HIERARCHICAL EXPERIMENT TO MEASURE THE VARIABILITY ASSOCIATED WITH THE GROUNDWATER SOURCE, SAMPLE PREPARATION AND INSTRUMENTATION

Sampling Method	Mean Activity (pCi/L)	Deviation (1σ) (pCi/L)	Mean % Relative Std. Dev.	% of Total Variance*
Direct Syringe	64,977	3,873	5.96	
Groundwater Source		1,087	1.67	7.9
Sample Prep. and Instrumentation		3,717	5.72	92.1
VOA Bottle	61,482	3,693	6.00	
Groundwater Source		0^+	0^+	0^+
Sample Preparation		2,752	4.47	55.5
Instrumentation		2,464	4.01	44.5

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* % of Total Variance = $\sigma_x^2/\sigma_T^2 \times 100$.

σ_x^2 = the variance due to the groundwater source, sample preparation or instrumentation.

σ_T^2 = the total variance including groundwater source, sample preparation and instrumentation.

+ Groundwater source deviation much less than those associated with sample preparation and instrumentation deviations, so it approaches zero in calculations.

because it can be transported through the mail without restriction and does not require special disposal. Recently, Packard (Sterling, VA) began manufacturing Opti-Fluor O, a cocktail designed for use in radon analysis, which can also be sent through the mail and discarded without special handling.

The results of the cocktail experiment (Fig. 102) indicated that all of the radium-226 standards approached secular equilibrium within 25 days, as predicted by theory. An ANOVA showed that the mean count rates for the mineral oil and Opti-Fluor O cocktails were not significantly different (α = 0.05 and 0.10), however, the mean count rate of the toluene-based cocktail was significantly less than both of these (α = 0.05 and 0.01). In addition, the %RSD associated with the toluene-based cocktail (3.22%) was greater than those of the mineral oil (0.90%) and Opti-Fluor O (1.34%). The H numbers (Appendix B) for all of the standards were low (Toluene = 15 ± 1.5, Mineral Oil = 25 ± 2.1, Opti-Fluor O = 17 ± 2.8) indicating that quenching was not a major factor for any of the cocktails. All three exhibited good two-phase separation from the water, although the mineral oil and Opti-Fluor O had small amounts of emulsion formation at the interface.

When the cocktail experiment was conducted using water samples collected at Derry and Lee, NH similar results were obtained (Table 28). At α = 0.05 and 0.01 (ANOVA), the toluene-based cocktail's count rates were significantly less than those of the mineral-based and Opti-Fluor O, which were not significantly different from each other (α = 0.05 and 0.10).

In all cases, the toluene-based cocktail yielded significantly fewer counts (~10%) than the mineral oil and Opti-Fluor O cocktails. The increased counts may have come from radium-226 contained in the small emulsions formed at the interface of the water and the Opti-Fluor O or mineral oil-based cocktails. To test this hypothesis, an experiment with the cocktail Instagel was performed. Since Instagel forms a single phase gel emulsion with the aqueous phase one would expect to obtain counts from radium-226 present in the water. Radium-226 standards made with Instagel (Fig. 103) had counts approximately 1.6 times greater than the other cocktails. The increased counts resulted from radium-226, lead-210, bismuth-210 and polonium-210 which emit alpha or beta particles, but are not normally soluble in the toluene, mineral oil and Opti-Fluor O cocktails. These radionuclides are miscible in Instagel and as a result, this cocktail is exposed to additional radioactive disintegrations compared to two-phase cocktails (Table 29) (Section 3).

The count rates obtained for the samples in the toluene-based cocktail taken at the Lee and Derry, NH sites were also approximately 10% lower than those taken in the mineral oil and Opti-Fluor O cocktails. In these samples, radium-226 activity was substantially lower (<10 pCi/L) than in the standards (588 pCi/L). Therefore, it is unlikely that the radium-226 activities in the samples were high enough in the mineral oil-based and Opti-Fluor O emulsions to account for the 10% higher count rate. Perhaps, it occurs because they have a higher counting efficiency than the toluene-based cocktail.

The standards containing toluene showed a significant increase in %RSD (α = 0.05 and 0.01, F test) over time compared with the other two cocktails

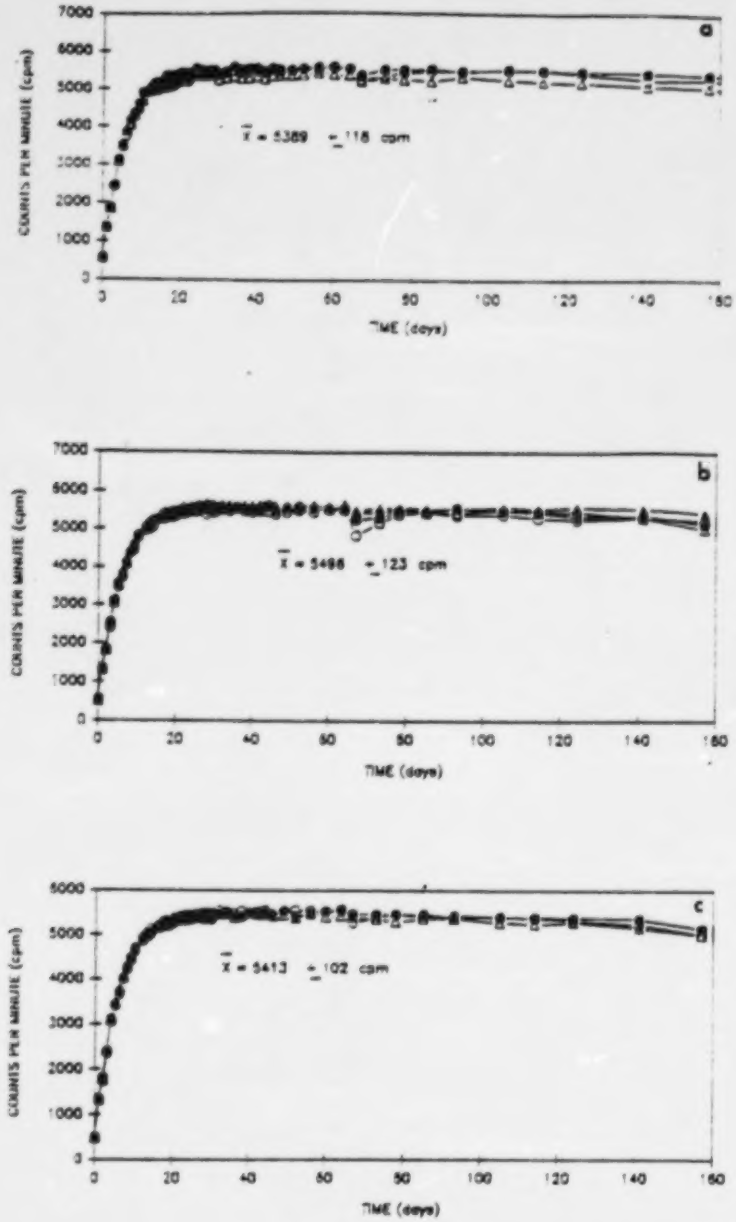

Figure 102. Cocktail experiment. Count rate (cpm) as a function of a time. (a) Toluene-based cocktail, (b) mineral oil-based cocktail, (c) Opti-fluor O.

TABLE 28. COCKTAIL EXPERIMENT COUNT RATES AT LEE AND DERRY, NH

Site	Scintillation Cocktail Toluene-Based (cpm)	Mineral-Oil Based (cpm)	Opti-Fluor O (cpm)
Lee	225±8*	244±9	248±7
Derry	3,028±68	3,426±58	3,483±42

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* S represents 1 standard deviation.

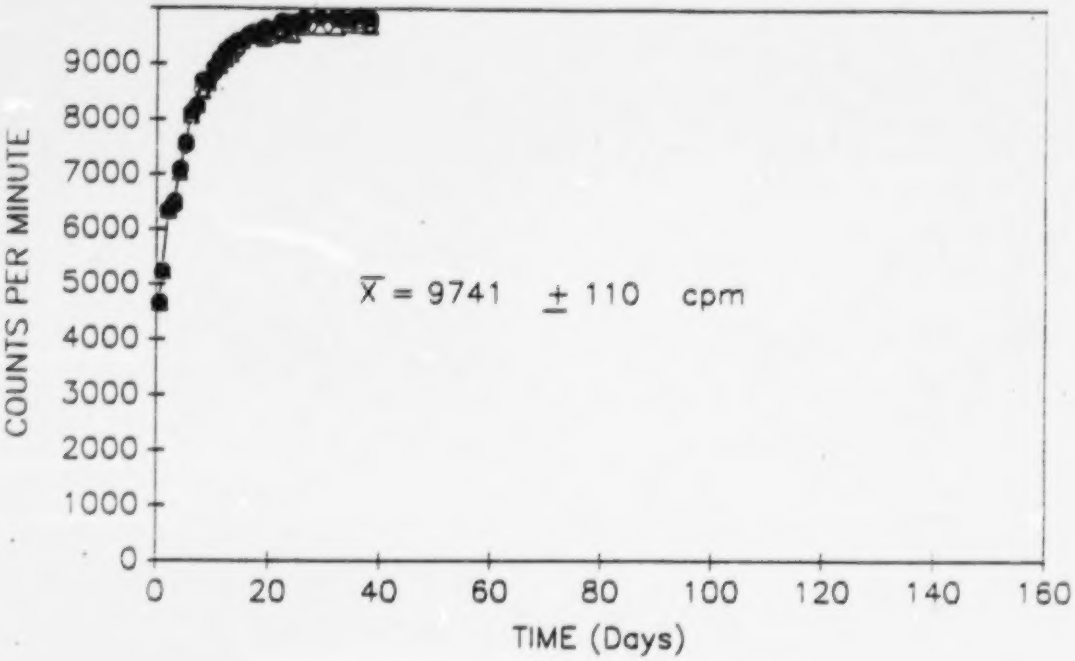

Figure 103. Cocktail experiment. Count rate (cpm) as a function of time for Instagel cocktail.

TABLE 29. RADIONUCLIDES PRESENT IN COCKTAILS YIELDING DISINTEGRATION

Instagel	Mineral Oil, Toluene, Opti-Fluor O
Radium-226	Radon-222
Radon-222	Polonium-218
Polonium-218	Lead-214
Lead-214	Bismuth-214
Bismuth-214	Polonium-214
Polonium-214	
Lead-210[+]	
Bismuth-210[+]	
Polonium-210[+]	

[*] Lead-210 low energy beta disintegrations will be counted at a 50-90% efficiency in a lower energy window (P.B. Hahn, USEPA Environmental Monitoring Systems Laboratory - Las Vegas, personal communication, 1989).

[+] These radionuclides are not in secular equilibrium with radium-226.

possibly because of volatilization of the cocktail. A 1-2 mL decrease in volume was observed in some of the scintillation vials stored in the laboratory over a period of 100 days in spite of the fact that they were tightly capped. This represents a loss of 10-20% of the cocktail indicating that toluene-based standards may have a limited shelf life. Others have reported that the mineral oil-based cocktail also has a limited life because by chemical reaction or some other means it degrades to a yellow color which in turn causes photodegradation and thus a reduction in light quality (R. Velten, USEPA Environmental Monitoring Support Laboratory - Cincinnati, 1987).

The choice of a cocktail may be influenced by other factors such as cost, disposal and mailing restrictions. Opti-Fluor O is the least expensive ($6.50/L), followed by the toluene-based ($8.50/L) and the mineral oil-based ($31.00/L) cocktails. The overall cost of the toluene-based cocktail is higher (~$12.00/L) when the disposal fees and special mailing requirements are included because toluene is an EPA listed hazardous material. Opti-Fluor O appears to provide the highest efficiency with the lowest variability and cost.

Extraction Experiment

The EPA procedure (1978) for analyzing radon in water requires that the capped vial containing the cocktail and water sample (or standard) be shaken for 1 min. This step is used to speed the extraction of the radon from the water into the cocktail while radionuclides, such as radium and uranium remain in the aqueous phase. The vial is held for 4 hr prior to counting to allow the short-lived progeny of radon to come to secular equilibrium so that a constant number of disintegrations will be occurring during analysis. The extraction experiment was conducted with each cocktail, to test the need for shaking, however the results from different cocktails should not be compared as they were prepared at different times.

The results for the standards with all of the cocktails had the expected increase in count rate over time as radon and its progeny came to secular equilibrium with radium-226 (Figures 104, 105 and 106). In all cases, there were only small differences in the count rates obtained for the standards regardless of whether or not they were shaken daily. If the extraction procedure was correct, the standards never shaken or shaken only on the first day should have had count rates approaching background. The data suggest that transfer of radon to the cocktail is continuous. The same pattern was observed for the samples taken at Lee and Derry (Table 30).

Though there were not large discrepancies between the count rates of the standards and samples as a result of shaking, there were statistically significant differences (Tables 31 and 32). However, there was no clear pattern emerging from these data. For the standards, the differences observed for each cocktail may be due to variations during preparation. The greatest contributors to variation in standards are pipetting, pipette rinsing and air stripping. During each of these steps, even with proper radiological techniques, small amounts of radium may be lost leading to variation in the count rates obtained. This is especially true for the bubbling of the standard which is difficult to regulate properly and consistently. Use of

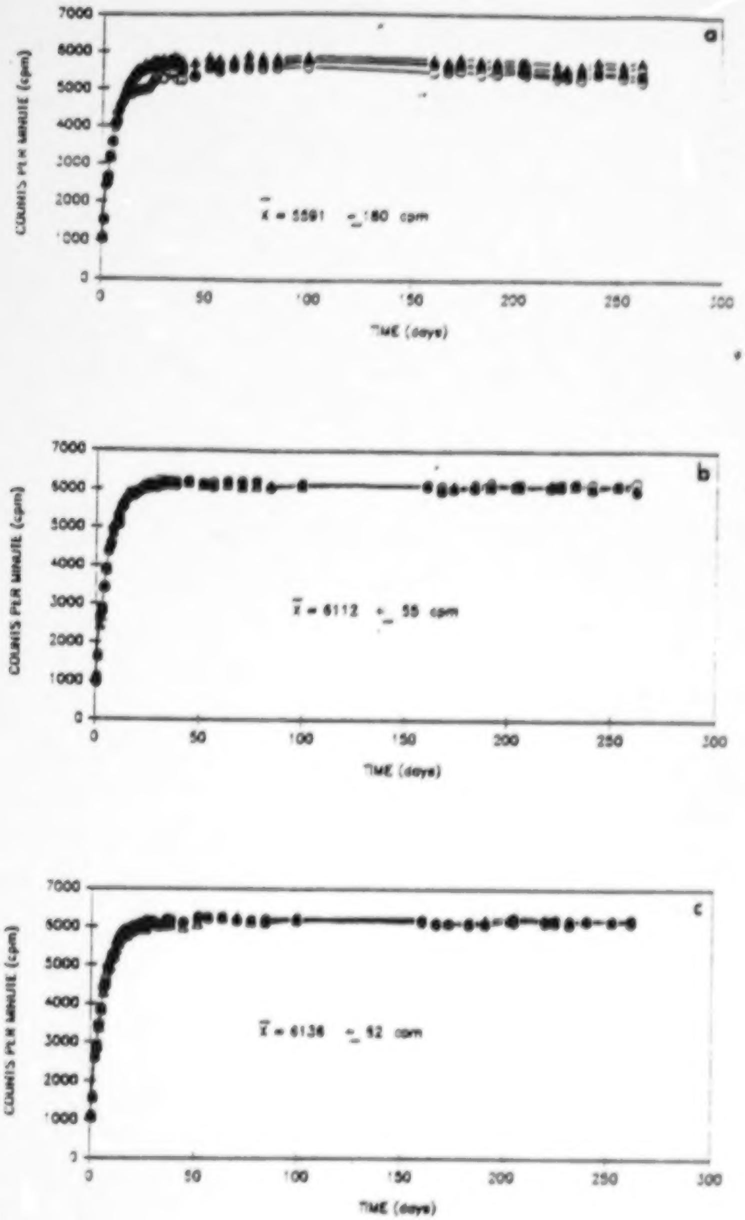

Figure 104. Extraction experiment. Count rate (cpm) for radium-226 standards using a toluene-based cocktail as a function of time. (a) Shaken daily, (b) shaken initially, (c) never shaken.

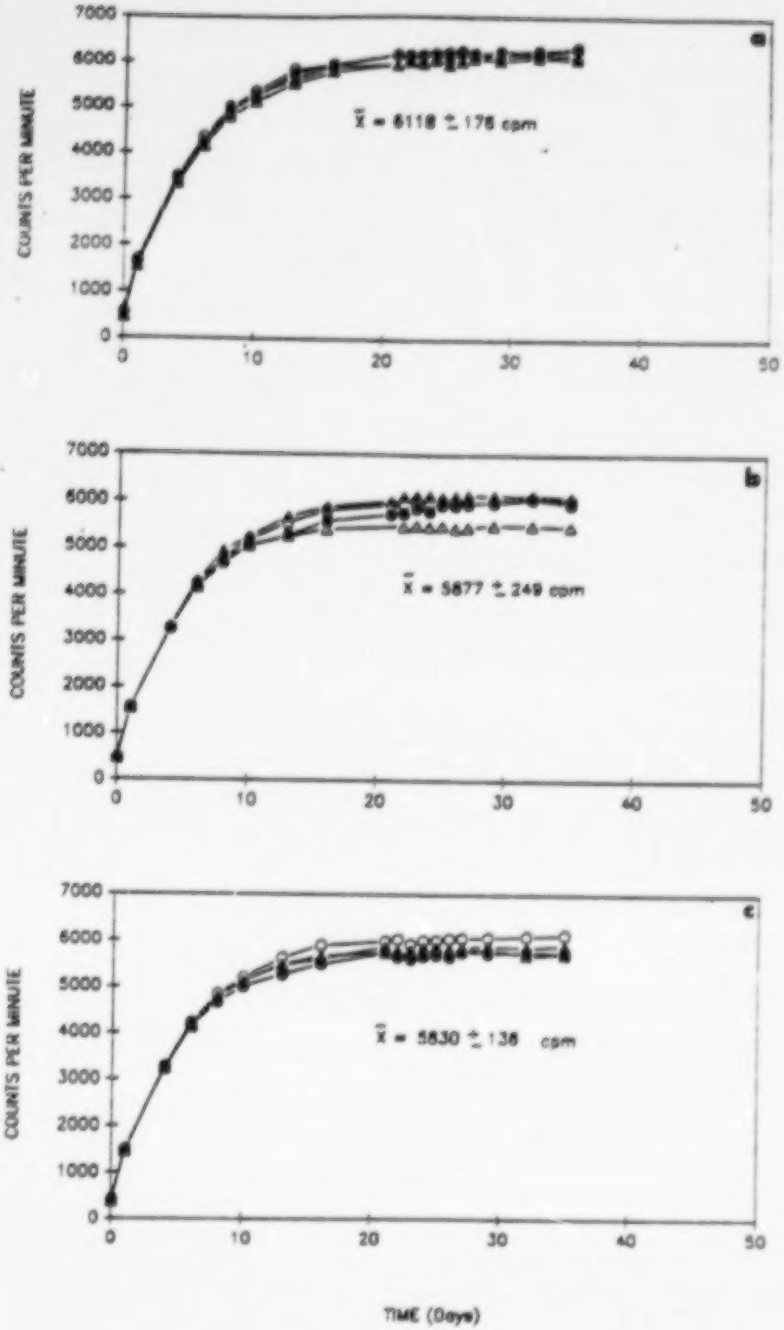

Figure 105. Extraction experiment. Count rate (cpm) for radium-226 using a mineral oil-based cocktail as a function of time. (a) Shaken daily, (b) shaken initially, (c) never shaken.

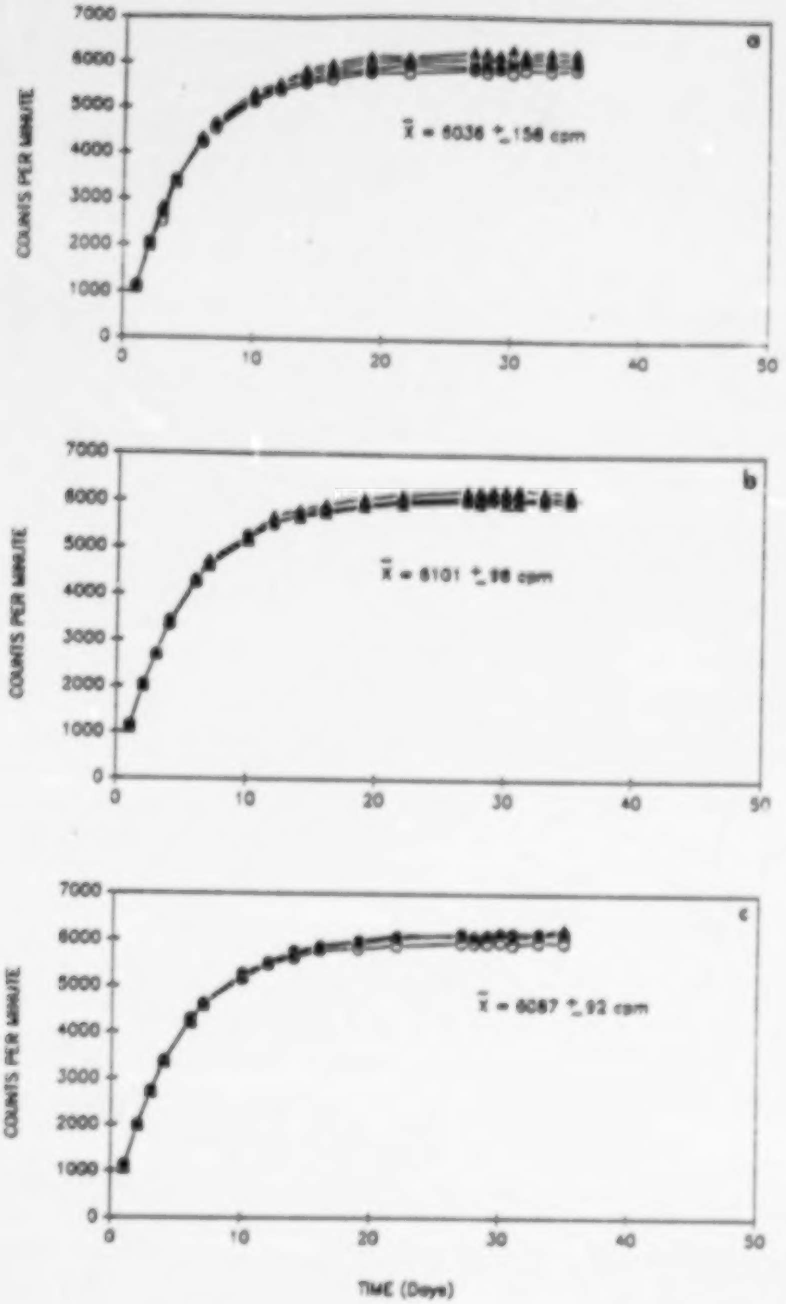

Figure 106. Extraction experiment. Count rate (cpm) for radium-226 using Opti-fluor O as a function of time. (a) Shaken daily, (b) shaken initially, (c) never shaken.

TABLE 30. MEAN COUNT RATES OF EXTRACTION EXPERIMENT AT LEE AND DERRY, NH[*]

Site	Toluene-Based (cpm)		Mineral Oil-Based (cpm)		Opti-Fluor O (cpm)	
	Never Shaken	Shaken	Never Shaken	Shaken	Never Shaken	Shaken
Lee,	274±12	289±6	240±7	288±10	254±14	283±11
Derry,	5,684±103	5,649±108	4,178±59	4,380±49	4,212±48	4,439±30

[*] Comparisons should not be made among cocktails as they were sampled on different days.

TABLE 31. STATISTICAL DIFFERENCES BASED ON MEAN COUNT RATES OF STANDARDS FOR EXTRACTION EXPERIMENT

Cocktail	Statistical Relationship[*]	Significance Level α
Toluene-Based	Shaken Daily ≡[+] Never Shaken	0.05
	Shaken Daily < Never Shaken	0.10
	Shaken Daily < Shaken Initially	0.05 & 0.01
	Never Shaken < Shaken Initially	0.05 & 0.01
Mineral Oil-Based	Shaken Initially ≡ Never Shaken	0.05 & 0.10
	Shaken Initially < Shaken Daily	0.05 & 0.01
	Never Shaken < Shaken Daily	0.05 & 0.01
Opti-Fluor O	Never Shaken ≡ Shaken Daily	0.05 & 0.10
	Shaken Daily ≡ Shaken Initially	0.05 & 0.10
	Never Shaken ≡ Shaken Initially	0.05 & 0.10

[*] As assessed by Scheffe analysis which was used because variances were not homogeneous.

[+] ≡: not significantly different at α specified.

TABLE 32 STATISTICAL DIFFERENCES BASED ON MEAN COUNT RATES OF SITES FOR EXTRACTION EXPERIMENT

Site	Cocktail	Statistical Relationship[*]	α
Lee, NH	Toluene-Based	Never Shaken < Shaken	0.05
		Never Shaken ≡[+] Shaken	0.02
	Mineral-Oil Based	Never Shaken < Shaken	0.05 & 0.01
	Opti-Fluor O	Never Shaken < Shaken	0.05 & 0.01
Derry, NH	Toluene-Based	Never Shaken ≡ Shaken	0.05 & 0.10
	Mineral Oil-Based	Never Shaken < Shaken	0.05 & 0.01
	Opti-Fluor O	Never Shaken < Shaken	0.05 & 0.01

[*] As assessed by Student's t test.

[+] ≡: not significantly different at α specified.

multiple point standard curves in the calculation of sample activities will help to overcome the effects of these variations in preparation. An interlaboratory QA/QC program is essential to insure that results obtained at different laboratories are comparable.

The data for the field samples collected at Lee and Derry indicate that, though extraction via shaking is not essential for radon transfer to the cocktail, it does yield significantly higher mean count rates (≤ 20%) for all cocktails. The extraction procedure for samples should be used to maximize the rate of transfer of radon to the cocktail phase.

The extraction experiment results have repercussions with respect to the calculation of the efficiency factor, E, (cpm/pCi) used to determine the radon activity in the samples (eq. 12). Extraction procedure (EPA, 1978) predicts that once radon-222 in the standards is in secular equilibrium with radium-226 in the aqueous phase, a known amount of radon is available to be extracted into the cocktail, but can only be transferred by shaking. In that case, the time between extraction (shaking) and analysis for the standards would determine the amount of radon and its progeny and hence, the number of disintegrations (dpm) occurring in the cocktail at the time of counting.

For example, if a standard containing 587 pCi of radium-226 were in secular equilibrium with radon-222, it would contain 587 pCi of radon. According to the procedure, all of the radon would remain in the aqueous phase until extraction. Immediately, after shaking nearly all of the 587 pCi of radon would be transferred to the cocktail. After 4 hr, the radon in the cocktail would be in secular equilibrium with its short-lived progeny. Based on the data from the extraction experiment, the count rate in the standard at 4 hr would be 5,400 and the net count rate would be 5340 (i.e., 5400 - 60 cpm for the blank). After 4 hrs, 2.98% of the original radon in the cocktail would decay away and not be regenerated from the aqueous phase, so only 569.5 pCi would be present when the analysis was performed. The efficiency factor is therefore 5,340 cpm/569.5 pCi = 9.38 cpm/pCi (equivalent to 5,340 cpm/1,264.3 dpm where 1 pCi = 2.22 dpm).

A similar calculation could be made if the standard were counted 24 hr after shaking. Based on the data from the extraction experiment, the count rate obtained would still be approximately 5,340, but the radon activity remaining in the cocktail according to the theory would have decayed to 489.7 pCi (83.4% of the original 587 pCi). Hence, the efficiency factor would be 10.90.

The effect of this change can be demonstrated by examining eq. 12. According to the extraction procedure, a 10 mL sample yielding a net 89 cpm (sample cpm - background cpm) and counted 12 hr after collection would contain 1,038 pCi/L assuming only 4 hr elapsed between shaking and counting of the standard, whereas the sample would be calculated to contain 894 pCi/L if the standard were counted 24 hr after shaking. However, in reality the actual radon activity would be 1,071 pCi/L based on the findings of the extraction experiment (i.e., the efficiency factor of the standard would remain 5,340 cpm/587 pCi or 9.10 cpm/pCi regardless of when the standard is counted). If the MCL for radon was set at 1,000 pCi/L, such a difference might be crucial.

Thus, if extraction principles are applied to standards, efficiency factors would be inflated leading to an underestimation of actual radon activities in the samples. This error would be especially critical for samples at or near the MCL.

Finally, it is important to analyze radium-226 standards (at secular equilibrium) during every sample run. As shown in Figures 104-106, the count rate obtained from standards analyzed on consecutive days using the same liquid scintillation counter may vary by as much as 5%. Preparation of a calibration curve based on radium-226 standards of various activities analyzed during each liquid scintillation counter run may also provide a better conversion of cpm to pCi. This is especially significant when samples with a variety of radon activities are analyzed.

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APPENDIX A

DAILY AVERAGE ANALYTICAL PRECISION AND ACCURACY

DIFFUSED BUBBLE

Analysis	pH		Alkalinity (mg/L as $CaCO_3$)		Turbidity (NTU)		Mn (mg/L)	Fe (mg/L)	Mn	Fe
Date	S_x^*	%RSD$^+$	S_x	%RSD	S_x	%RSD	S_x	S_x	%REC**	%REC
7-28-88	0.03	<1	1.27	2	5.3	40	-	-	-	-
8-24-88	0.04	<1	0.63	1	0.01	8	-	-	-	-
8-25-88	0.02	<1	0.00	0	0.09	13	-	-	-	-
8-26-88	0.01	<1	0.96	3	0.06	2	-	-	-	-
8-29-88	0.01	<1	7.00	8	0.02	11	-	-	-	-
8-30-88	0.02	<1	0.95	3	0.08	1	-	-	-	-
9-1-88	0.03	<1	2.23	4	0.67	25	-	-	-	-
9-2-88	0.01	<1	1.20	2	0.01	4	-	-	-	-
9-15-88	0.05	1	2.33	4	0.03	2	-	-	-	-
9-11-89	-	-	-	-	-	-	0.01	0.02	110	109
PACKED TOWER										
8-4-87	0.01	<1	0.00	0	-	-	-	-	-	-
8-7-87	0.01	<1	0.17	1	0.00	<1	-	-	-	-
8-10-87	0.04	<1	0.27	1	0.01	17	-	-	-	-
8-12-87	0.01	<1	0.17	<1	0.01	4	-	-	-	-
8-14-87	0.02	<1	0.54	2	0.01	21	-	-	-	-
8-18-87	0.04	<1	0.54	2	0.05	51	-	-	-	-
8-20-87	0.00	<1	0.00	<1	0.00	0	-	-	-	-
9-1-87	0.10	1	0.06	<1	0.00	0	-	-	-	-
9-3-87	0.02	<1	0.00	<1	0.00	1.2	-	-	-	-
9-4-87	0.07	1	1.43	6	0.02	38	-	-	-	-
9-9-87	0.01	<1	0.00	<1	0.01	25	-	-	-	-
9-10-87	0.02	<1	0.18	<1	0.00	0	-	-	-	-
9-11-87	0.05	<1	0.17	<1	0.00	0	-	-	-	-

(continued)

PACKED TOWER (continued)

Analysis Date	pH S_x^*	pH %RSD$^+$	Alkalinity (mg/L as $CaCO_3$) S_x	Alkalinity %RSD	Turbidity (NTU) S_x	Turbidity %RSD	Mn (mg/L) S_x	Fe (mg/L) S_x	Mn %REC**	Fe %REC
1-30-88	0.02	<1	0.00	<1	0.06	18	-	-	-	-
2-3-88	0.02	<1	0.23	<1	0.01	8.4	-	-	-	-
2-5-88	0.02	<1	0.37	2	0.01	12	-	-	-	-
2-21-88	-	-	-	-	-	-	0.00	0.00	124	113
2-24-88	-	-	-	-	-	-	0.00	0.00	84	88
6-14-88	-	-	-	-	-	-	0.01	0.01	107	98
6-17-88	-	-	-	-	-	-	0.01	0.01	108	95

GAC SYSTEMS

Analysis Date	pH S_x^*	pH %RSD$^+$	Alkalinity (mg/L as $CaCO_3$) S_x	Alkalinity %RSD	Turbidity (NTU) S_x	Turbidity %RSD	Mn (mg/L) S_x	Fe (mg/L) S_x	Mn %REC**	Fe %REC
10-9-86	-	-	2.12	9	0.00	0	-	-	-	-
11-30-86	-	-	0.35	1	0.26	14	-	-	-	-
12-4-86	0.01	<1	0.22	1	0.10	3	-	-	-	-
12-11-86	0.02	<1	0.62	3	0.04	8	-	-	-	-
12-18-86	0.04	<1	0.15	1	0.01	13	-	-	-	-
1-7-87	0.01	<1	0.18	1	0.04	13	-	-	-	-
1-16-87	0.02	<1	0.27	1	0.05	18	-	-	-	-
1-24-87	0.02	<1	0.09	<1	0.00	0	-	-	-	-
1-29-87	0.01	<1	0.29	1	0.01	6	-	-	-	-
2-9-87	0.01	<1	0.29	1	0.02	1	-	-	-	-
2-11-87	0.02	<1	0.59	2	0.01	10	-	-	-	-
2-18-87	0.02	<1	0.71	3	0.01	2	-	-	-	-
2-25-87	0.05	1	0.35	2	0.01	15	-	-	-	-
3-4-87	0.02	<1	0.59	2	0.10	18	-	-	-	-
3-11-87	0.03	<1	0.41	1	0.03	13	-	-	-	-
3-13-87	-	-	-	-	-	-	0.01	0.01	136	205
3-16-87	-	-	-	-	-	-	0.01	0.01	110	142
3-17-87	0.00	<1	0.00	0	0.00	0	-	-	-	-
4-14-87	0.04	1	0.00	0	0.01	17	-	-	-	-
5-13-87	0.02	<1	0.00	0	0.01	15	-	-	-	-
6-11-87	0.05	1	0.18	1	0.00	0	-	-	-	-
6-15-87	-	-	-	-	-	-	0.00	0.01	75	90
6-16-87	-	-	-	-	-	-	0.00	0.01	83	90
6-17-87	-	-	-	-	-	-	0.00	0.01	97	90
6-21-87	-	-	-	-	-	-	0.01	0.01	81	106
6-25-87	-	-	-	-	-	-	0.00	0.01	89	80

(continued)

GAC SYSTEMS (continued)

Analysis	pH		Alkalinity (mg/L as $CaCO_3$)		Turbidity (NTU)		Mn (mg/L)	Fe (mg/L)	Mn	Fe
Date	S_x*	%RSD+	S_x	%RSD	S_x	%RSD	S_x	S_x	%REC**	%REC
7-1-87	0.01	<1	0.25	1	0.00	0	-	-	-	-
7-2-87	0.01	<1	0.53	2	0.00	0	-	-	-	-
7-7-87	0.01	<1	0.14	2	0.00	0	-	-	-	-
7-16-87	0.04	1	0.35	2	0.00	0	-	-	-	-
8-4-87	0.05	1	0.35	2	0.00	0	-	-	-	-
8-6-87	-	-	-	-	-	-	0.01	0.01	98	102
8-11-87	-	-	-	-	-	-	0.00	0.01	75	65
8-13-87	0.04	1	0.36	2	0.00	0	0.00	0.00	-	-
9-14-87	0.04	1	0.00	0	0.00	0	-	-	-	-
9-17-87	0.08	1	0.00	0	0.00	0	-	-	-	-
1-28-88	0.14	2	0.40	1	0.01	15	-	-	-	-
1-29-88	0.03	4	0.50	1	0.01	11	-	-	-	-

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* S_x is the average standard deviation.

+ %RSD is the average percent relative standard deviation.

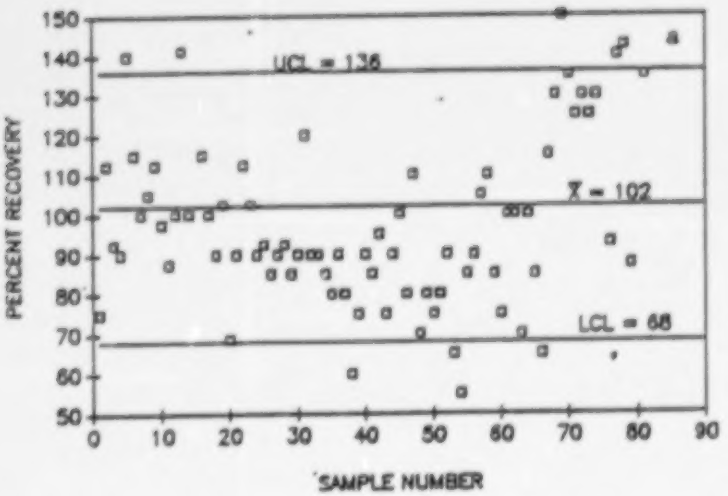
** %REC is the average percent recovery.

RADON

The analytical precision, as % relative standard deviation (%RSD), was calculated several times during the course of the project. This was done by counting different blanks and standards three consecutive times in the liquid scintillation counter. The % RSD data are shown below:

Sample	$\bar{X}$ (pCi/L)	S_x (pCi/L)	% RSD
Blank	83.2	3.3	3.9
Blank	99	4.4	4.4
Blank	98	4.0	4.1
Blank	57	2.2	3.9
Blank	56	3.6	6.4
Standard	3,028	168.4	5.6
Standard	4,019	230	5.7
Standard	4,921	58	1.2
Standard	5,195	35	0.7
Standard	4,994	126	2.5
Standard	4,902	159	3.2
Standard	4,902	159	3.2
Standard	4,921	58	1.2

The percent % RSD, as shown in Section 8, for the scintillation counter is usually < 6%. Duplicate samples were also run every tenth sample, however, these samples were taken consecutively and therefore, reflect variability in the source as well as the analysis.



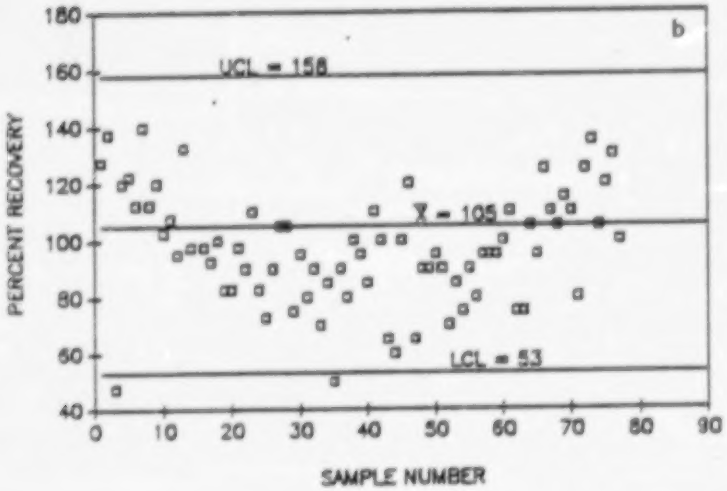
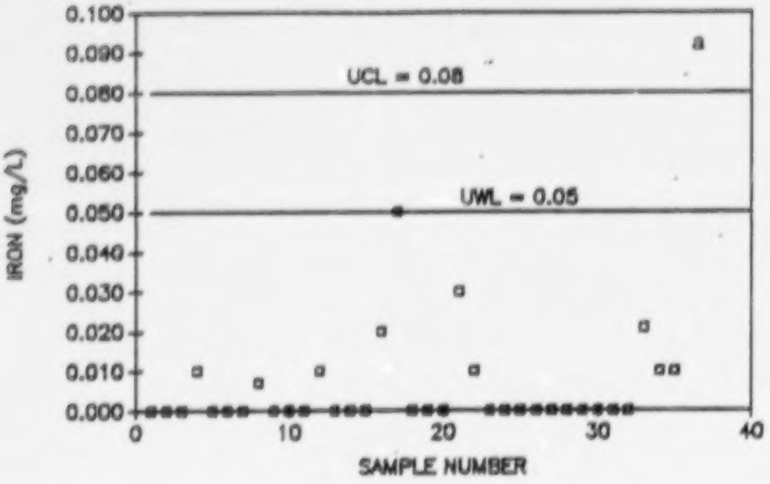


Figure A1. Mont Vernon, NH GAC system. Control charts for accuracy. (a) Iron and (b) manganese.



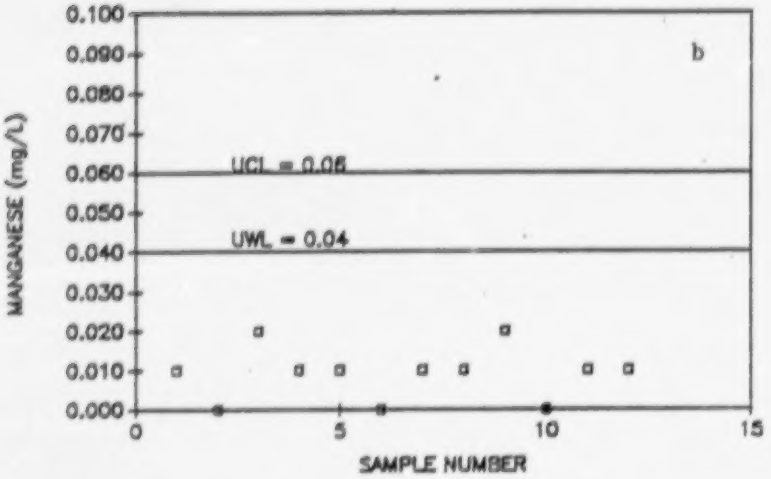


Figure A2. Mont Vernon, NH GAC system. Control charts for precision. (a) Iron and (b) manganese.

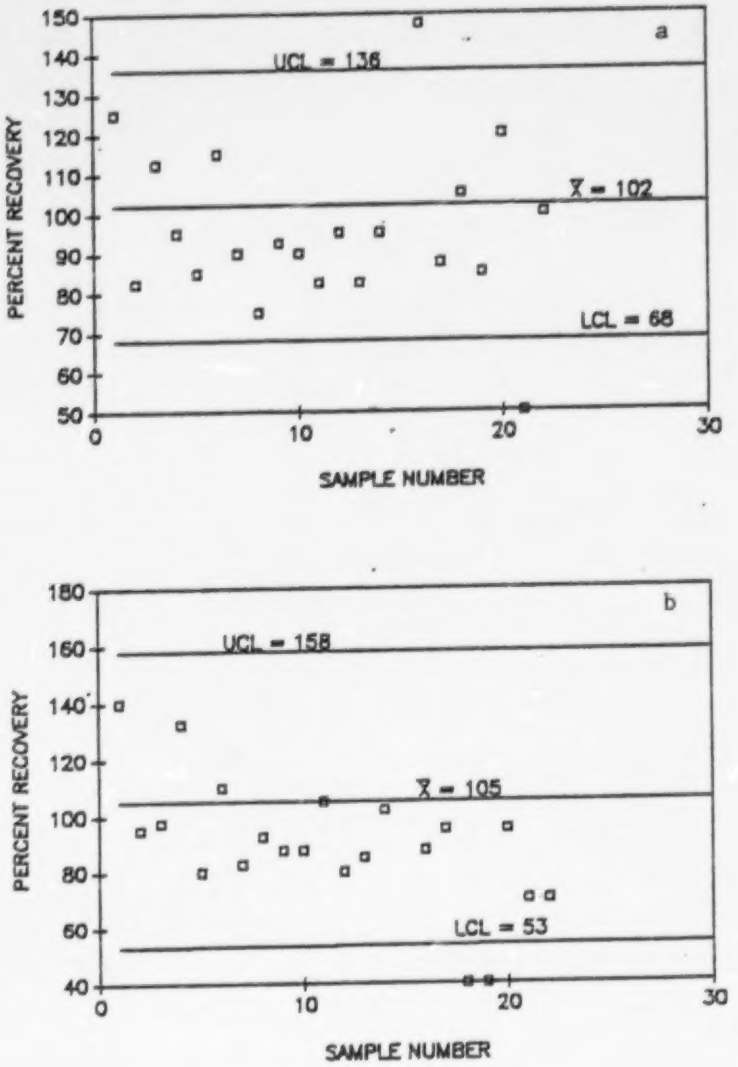
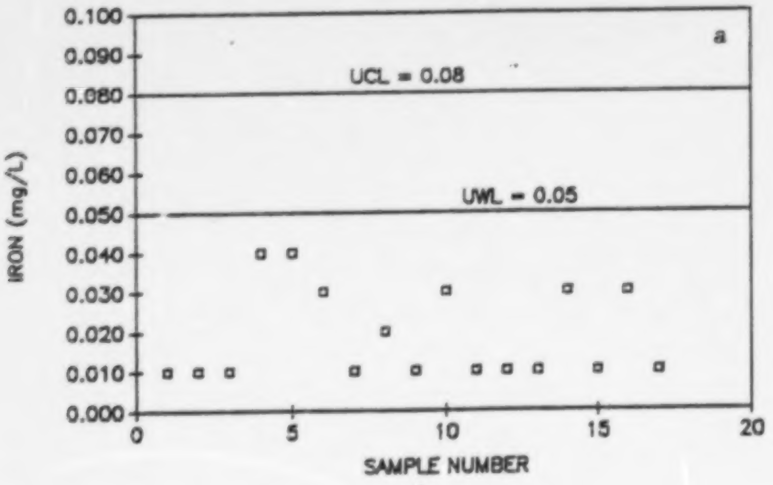


Figure A3. Amherst, NH GAC system. Control charts for accuracy. (a) Iron and (b) manganese.



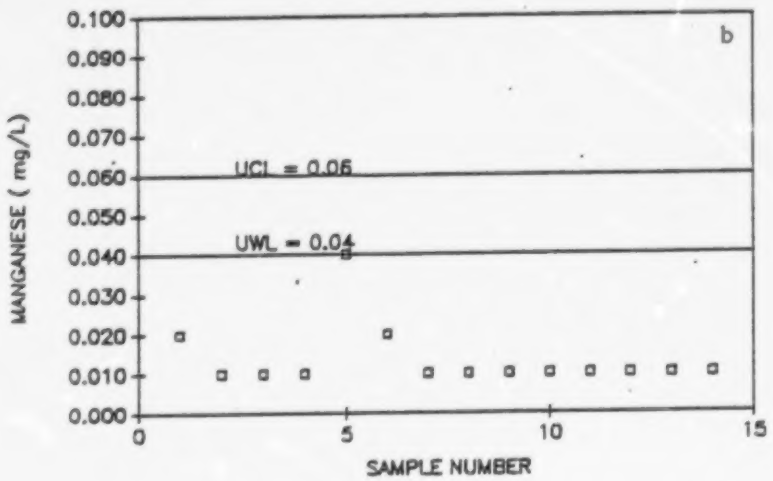


Figure A4. Amherst, NH GAC system. Control charts for precision. (a) Iron and (b) manganese.

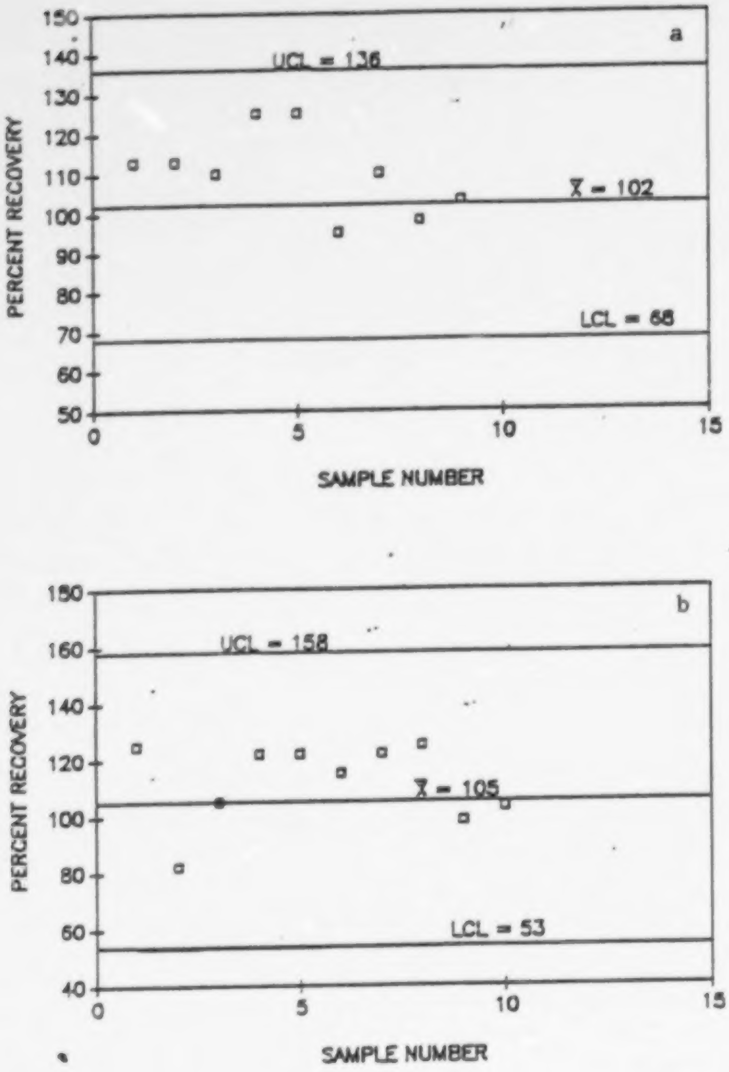
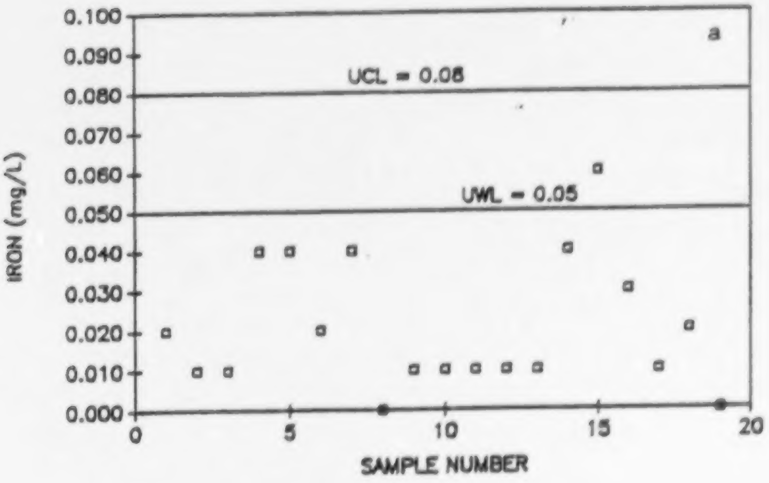


Figure A5. Diffused bubble aeration. Control charts for accuracy. (a) Iron and (b) manganese.



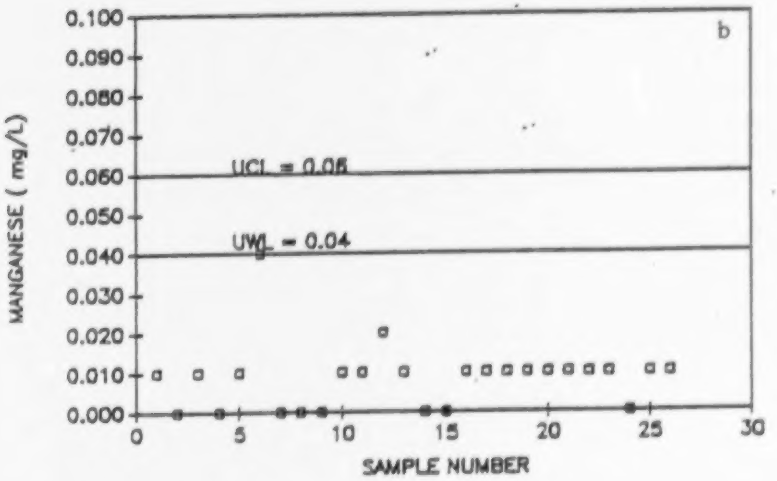


Figure A6. Diffused bubble aeration. Control charts for precision. (a) Iron and (b) manganese.

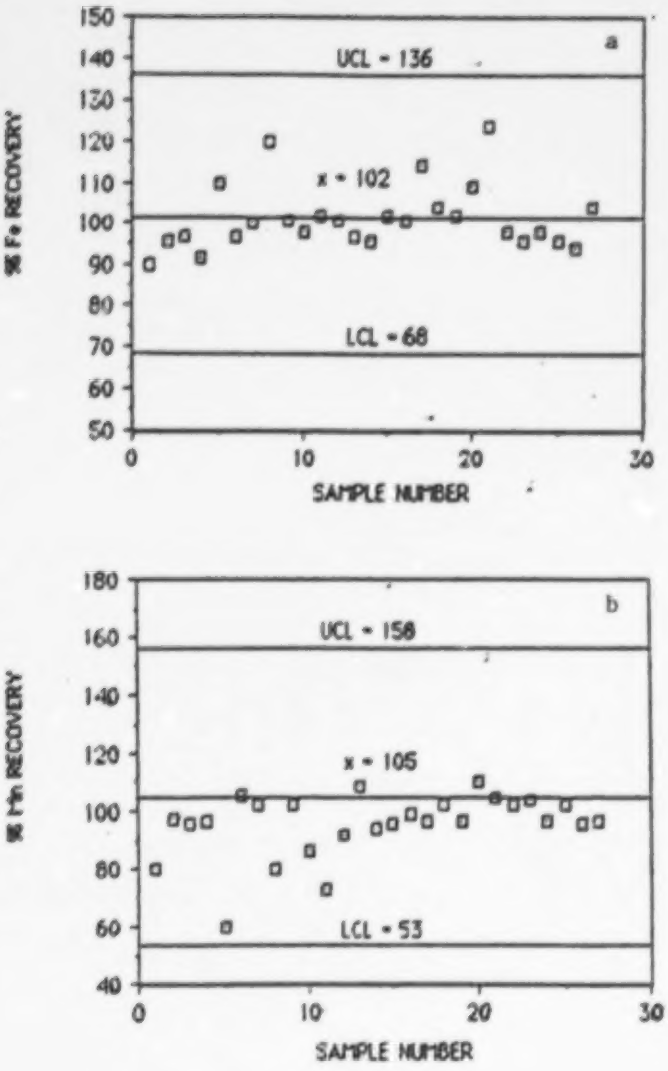
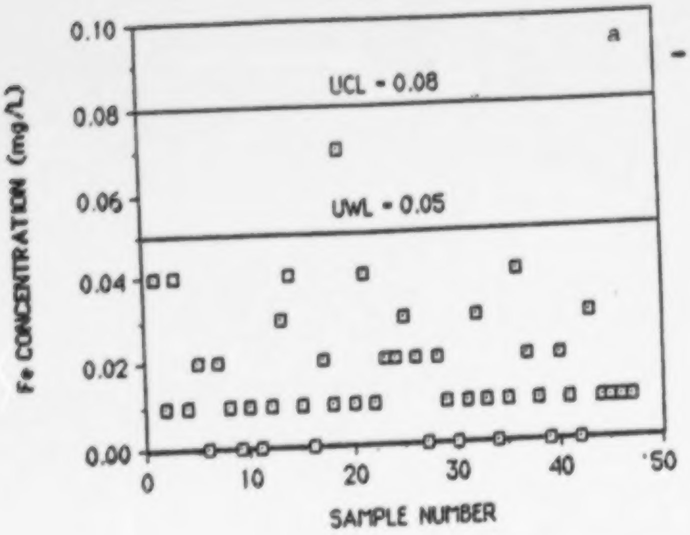


Figure A7. Packed tower aeration. Control charts for accuracy. (a) Iron and (b) manganese.



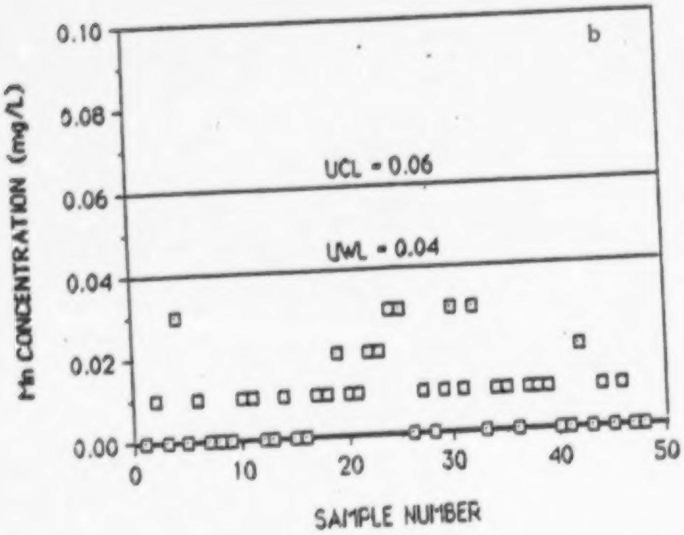


Figure A8. Packed tower aeration. Control charts for precision. (a) Iron and (b) manganese.

APPENDIX B

SUMMARY OF LIQUID SCINTILLATION COUNTER PROGRAM

The program outlined below was obtained from the operations manual for the Beckman (Fullerton, CA) LS 7000 liquid scintillation counter used in this research project. It is library program #6 for this instrument.

Unmodified Count Time 10 min

Channels	Counted	Lower Limit	Upper Limit
1	2.00σ%*	0	397
2	2.00σ%*	397	940

Sample Channels Ratio	No
H Number Calibration**	1 time (in channels 1 & 2)
Automatic Quench Compensation	Yes
Data Printout Units	cpm

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*2.00σ% = $200/\sqrt{N}$ where N = total number of counts obtained at the time of calculation.

**The H-number quantifies quench and indicates the effect of interferences on the counting efficiency. Automatic quench compensation uses an internal cesium-137 source to generate an H-number for each sample, which the liquid scintillation counter uses to adjust the count rate generated in correspondence to the quenching.

APPENDIX C

METHODS OF CALCULATION OF LEAD-210 ADSORPTION

I. Calculation of Lead-210 activity from gamma counter data using channels 89-96.

$$A' = \frac{\text{Net Peak Counts}}{(t)\ (m)\ (E)\ (I_g)}$$

where A' = activity (dpm/g GAC dry weight)

Net Peak Counts = (Adjusted Peak Counts - Adjusted Compton Counts)

Adjusted Peak Counts = (Sample Peak Counts - Background Peak Counts)

Adjusted Compton Counts = (Sample Compton Counts - Background Compton Counts)

t = time sample counted (min)

m = mass of sample (g)

E = efficiency factor based on counts obtained from Lead-210 standard sorbed to equivalent volume of virgin GAC (cpm/dpm)

I_g = 0.04 = amount of Lead-210 emissions which are gamma radiation

$$A = A' \cdot \frac{1\ \text{pCi}}{2.22\text{dpm}} \cdot \frac{1{,}000\ \text{g}}{\text{kg}}$$

where $A = \frac{\text{pCi Lead-210 measured}}{\text{kg GAC (dry weight)}}$

II. Calculation of theoretical Lead-210 accumulated on the GAC.

Assumptions:

(i) All radon-222 removed by the GAC was completed retained.

(ii) All lead-210 accumulation originates from decay of the radon-222 progeny. Any direct adsorption of lead-210 from the water supply or lead-210 resulting from the decay of other adsorbed species such as radium-226 was considered negligible.

(iii) The measured water flowrate and radon removed by a given volume of GAC were constant over a sampling interval.

A. Calculation of pCi of Radon-222 adsorbed in a given volume of GAC during the entire operating period (up to time of coring)

$$A_{i_{Rn}} = Q_{v_i}(C_{inf_i} - C_{eff_i})t_i$$

where $A_{i_{Rn}}$ = pCi radon adsorbed during sampling interval i

Q_{v_i} = water flowrate during sampling interval i (L/d)

C_{inf_i} = measured influent radon into volume of GAC during sampling period i (pCi/L)

C_{eff_i} = measured effluent radon out of volume of GAC during sampling period i (pCi/L)

$(C_{inf_i} - C_{eff_i})$ = net change in radon activity through the volume of GAC (pCi/L)

t_i = length of time of sampling interval (d)

$$A_{T_{Rn}} = \sum_{i=1}^{n} A_{i_{Rn}}$$

where $A_{T_{Rn}}$ = pCi of radon adsorbed to a given volume of GAC during the entire operating period (up to time of coring)

B. Calculation of mass of Radon-222 adsorbed to a given volume of GAC

$$M_{Rn} = A_{T_{Rn}} \times \frac{6.50 \times 10^{-18}\text{g Rn}}{\text{pCi Rn}}$$

where M_{Rn} = g of radon adsorbed to a given volume of GAC during the operating period (up to the time of coring)

$$\frac{6.50 \times 10^{-18}\text{g Rn}}{\text{pCi Rn}} = \frac{2.22\text{ dpm}}{1\text{ pCi}} \times \frac{1}{\lambda_{Rn}} \times \frac{\text{mole Rn}}{6.023 \times 10^{23}\text{ atoms}} \times \frac{222\text{gRn}}{\text{mole Rn}}$$

$$\lambda_{Rn} = \frac{\ln 2}{t_{1/2_{Rn}}} = \frac{0.693}{(3.82\text{d})(24\text{hr/d})(60\text{min/hr})} = 1.26 \times 10^{-4}\text{ min}^{-1}$$

where λ_{Rn} = number of radon atoms disintegrating per total radon atoms present per minute

$t_{1/2_{Rn}}$ = half-life of Radon-222 = 3.82d

C. Calculation of atoms of Radon-222 absorbed to a given volume of GAC.

$$N_{Rn} = M_{Rn} \times \frac{\text{mole Rn}}{222\text{gRn}} \times \frac{6.023 \times 10^{23}\ \text{atoms Rn}}{\text{mole Rn}}$$

where N_{Rn} = number of atoms of radon adsorbed to a given volume of GAC

D. Calculation of mass of Lead-210 (Pb) adsorbed to a given volume of GAC

$$N_{Pb} = N_{Rn}$$

where N_{Pb} = number of atoms of Lead-210 adsorbed to a given volume of GAC

$$M_{Pb} = N_{Pb} \times \frac{\text{mole Pb}}{6.023 \times 10^{23}\ \text{atoms}} \times \frac{210\text{g Pb}}{\text{mole Pb}}$$

where M_{Pb} = g of Lead-210 adsorbed to a given volume of of GAC

E. Calculation of pCi of Lead-210 (Pb) adsorbed to a given volume of GAC

$$A_{T_{Pb}} = M_{Pb} \times \frac{\text{pCi Pb}}{1.24 \times 10^{-14}\text{g Pb}}$$

$$\frac{\text{pCi Pb}}{1.24 \times 10^{-14}\text{g Pb}} = \frac{\text{pCi}}{2.22\ \text{dpm}} \times \lambda_{Pb} \times \frac{6.023 \times 10^{23}\ \text{atoms Pb}}{\text{mole Pb}} \times \frac{\text{mole Pb}}{210\text{g Pb}}$$

where $A_{T_{Pb}}$ = pCi Lead-210 adsorbed to a given volume of GAC

$$\lambda_{Pb} = \frac{\ln 2}{t_{1/2_{Pb}}} = \frac{0.693}{(21\text{yr})(\frac{365\text{d}}{\text{yr}})(\frac{24\text{hr}}{\text{d}})(\frac{60\text{min}}{\text{hr}})} = 6.28 \times 10^{-8}\ \text{min}^{-1}$$

where λ_{Pb} = number of Lead-210 atoms disintegrating per total Lead-210 atoms present per minute

$t_{1/2_{Pb}}$ = half-life of Lead-210 = 21 yr

F. Calculation of pCi of Lead-210 per mass of GAC:

$$\frac{\text{pCi Lead-210}}{\text{kg}} = \frac{A_{T_{Pb}}}{x}$$

where x = kg (dry weight) of GAC per given volume